

Lisbon, 3rd July 2020

Dear Editor Luc Beaufort,

We are submitting the revised version of the manuscript " Coccolithophore productivity at the western Iberian Margin during the middle Pleistocene (310 – 455 ka) – evidence from coccolith Sr/Ca data " [cp-2019-131] in which we address comments of the two reviewers.

We greatly appreciate the reviewers' comments and have incorporated them to improve this latest version. Suggestions by reviewer Tom Dunkley Jones resulted to major changes in the discussion with subchapters 5.2 and 5.3 being mostly re-written or newly added. The spectral analyses results and related text and figures have been omitted from the revised manuscript.

You will find below a detailed reply (in green) to all the points raised along with the specification of the revisions that have been made and arguments that we used in considering the comments. A tracked-changes version of this manuscript is also at the end of this rebuttal letter.

Once more, thank you very much for your understanding throughout the revision process and under such a challenging pandemic situation.

Best regards, on behalf of all co-authors,
Catarina Cavaleiro

Response to comments from the editors and reviewers:

-Reviewer 1

This is a very nice piece of work, presenting new high-resolution coccolith Sr/Ca records from the mid Pleistocene of the Iberian Margin. Coccolith Sr/Ca is an underused but potentially powerful proxy for understanding the cellular growth and calcification rates of the dominant marine calcifying phytoplankton, the coccolithophore algae.

Of particular value in this manuscript, is the integration of these records of growth rates, with other records of export flux to sediments of both the organic (alkenones) and inorganic (coccolith) carbon fixed by these phytoplankton. Together these allow a nuanced interpretation – including the coupling / decoupling - of surface ocean growth conditions and aggregated net export. I would recommend publication with revisions. In particular I would recommend shortening the manuscript and focusing on the strongest signals within the data in order to generate the impact that this work deserves.

Comments (in order of the text):

Line 17 – instead of “climate models” better to say “Earth System Models” as the common understanding of a “climate model” is one that doesn't include biogeochemistry.

And again at Line 48 (and anywhere else) – “climate models” including coccolithophore productivity – better to talk about Earth System and/or biogeochemical and/or carbon cycle models.

We thank the reviewer for this comment and changed this accordingly.

Line 18 – “coccolithophore paleoproductivity past reconstructions” doesn't make sense

We thank the reviewer for this comment and have changed the previous sentence to “the reconstruction of coccolithophore paleoproductivity mostly relied on proxies dependent on accumulation and sedimentation rates, and preservation conditions.”

Line 23 – define SST at first usage

We thank the reviewer for this comment and this was changed accordingly.

Lines 38-42 long sentence that jams together two concepts – split.

We thank the reviewer for this comment and since the introduction was re-structured, this long sentence was deleted.

INTRODUCTION

In both the introduction and the discussion, I feel the absence of a clearly articulated question – what is the “knowledge gap” and how does this paper address that gap?

There are suggestions of problems in the representation of coccolithophore production and export in biogeochemical models, but no sense of what the specifics of these are, or how they might be addressed by this study. When I first read this section, I was not convinced that models could be informed by new coccolith Sr/Ca records (but see below). Then there is the time period studied – there is a general overview of the significant changes going on through this interval, but the rationale for looking at the coccolithophore response is so broad that it loses meaning: “to evaluate this phytoplankton group’s behaviour and gain a better understanding of its response to climate conditions during glacials, interglacials, deglaciations and the transition from interglacial to glacial conditions, at both orbital and sub-orbital time scales.”

(line 85 & on). Or: “We aim to characterise long-term changes in coccolithophore productivity in such a system, where their behaviour in the past remains unknown.”

(line 74) or: “. . .and evaluate the main factors influencing coccolithophore productivity.”

(line 77) I would really like specifics of: 1) the dynamics / processes that you seek to investigate and 2) why these intervals.

I think part of your struggle is related to: 1) setting up the Sr/Ca as something that is a “better” measure of coccolithophore “productivity” than other approaches – e.g. NAR / alkenone accumulation; and 2) equating coccolith Sr/Ca with quite a loose concept of “productivity”.

Together these end up setting off your “productivity” records “against” one another, rather than being mutually informative about different components of the growth – export – accumulation system. This seems to lead to a discussion which is phrased in terms of “mismatch” rather than one that allows the complexity of the system response to be seen, because, you’ve got the advantage of multi-proxy data covering different aspects of the same system.

My recommendation is that you recast this introduction somewhere along these lines:

1) There are significant uncertainties about the complex interactions between coccolithophore growth rates, nutrient dynamics, seasonality, export (carbonate / organic carbon), dissolution and final accumulation / burial rates. These uncertainties make modelling the responses of this system to modern environmental change problematic.

2) These questions can only be addressed with: a) records that test the dynamic response of various components of the system over a reasonable range of change (i.e. palaeo records); and b) through multi-proxy studies of growth environment (Sr/Ca) and export of both organic (alkenones) and inorganic (NAR) carbon.

3) Then make the argument for the particular time period studied providing the chance to test a range of particular environmental conditions – and make your introduction to the time period outline what these might be – e.g. upwelling, seasonality, temperature...

With this set-up, hopefully you’ll then be able to circle round in the discussion and answer these questions.

We thank the reviewer for this comment and we have changed the introduction accordingly. Please check the tracked changes document.

Sr/Ca VARIATIONS AND ASSEMBLAGE CHANGES

Line 68 – 69: I’d like some more justification for the assumption that “assemblage changes don’t matter”. My reading of the Fink et al. 2010 paper was that the abundance of the larger *Calcidiscus leptoporus* did have a significant impact on CF Sr/Ca.

I’m also suspicious of using the logic that in some instances in the modern oceans CF Sr/Ca changes coincide with productivity changes, therefore, it must be productivity, when coincident

assemblage changes haven't been properly considered. Given expertise of Baumann and Stoll, I would like to see some more justification of this point, that CF Sr/Ca are really dominated by changes in growth rate, rather than assemblages, especially with respect to *Calcidiscus*, which I suspect can contribute strongly to some records that show large Sr/Ca variations.

We thank the reviewer for this comment. In our study we focus on the Mid-Brunhes interval exactly because during that time the coccolithophore flora was dominated by *gephyrocapsids* (97 % average, 60 % of *Gephyrocapsa caribbeanica*) with very low abundances of *Calcidiscus leptoporus* and *Helicosphaera* sp.. So contrary to the modern assemblages studied by Fink et al. (2010), it seems unlikely that changes in the range of the latter two's average relative or absolute abundance would have a significant effect on the coccolith fraction Sr/Ca ratio in our study. Please also check the reply to the next comment and to a similar comment from reviewer #2. We also believe that indeed the introduction stated too firmly that changes in the coccolith assemblages do not influence the coccolith fraction Sr/Ca ratio based on the papers from Barker et al., 2006 and Stoll et al., 2002a (see manuscript for references). Fink et al. 2010 present divergent data and we added a sentence to highlight the importance of considering the influence of assemblages' changes on the coccolith fraction Sr/Ca ratio before interpreting it as a paleoproductivity reconstruction proxy. The respective paragraph has now been moved to method subchapter 3.3.

Later – Line 179 – you talk about being in the *Gephyrocapsa* acme and that this makes assemblage variability less of an issue for CF Sr/Ca, but this somewhat admits that assemblage change can be an issue under other circumstances. A clear delineation – even without hard and fast data / rules – between when assemblage changes are likely and not-likely a problem with CF Sr/Ca would be better than trying to imply there is no issue. Please be precise with the logic and transparent with the reader as to if and where assemblage change might be playing a significant role.

The reviewer is correct when stating that indeed changes in the coccolith assemblage might bias the interpretation of the coccolith fraction Sr/Ca proxy for coccolithophore productivity. However, we believe that to delineate a threshold above which changes in coccolith assemblage would significantly bias the coccolith fraction Sr/Ca ratio is out of the scope of this research. As mentioned previously, in our research it is very unlikely that the changes in *Calcidiscus leptoporus* abundance could bias the CF Sr/Ca results, given the *gephyrocapsids* dominance. Plus, statistically, we find that there is no significant relationship between the relative abundance of *Calcidiscus* and the coccolith fraction Sr/Ca ratio or with the coccolithophore productivity proxy (see correlation charts and Pearson's correlation results below – Figure 1 and Table 1).

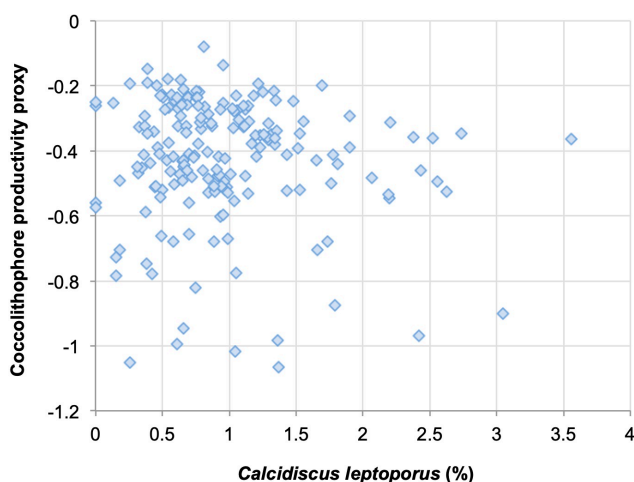


Figure 1

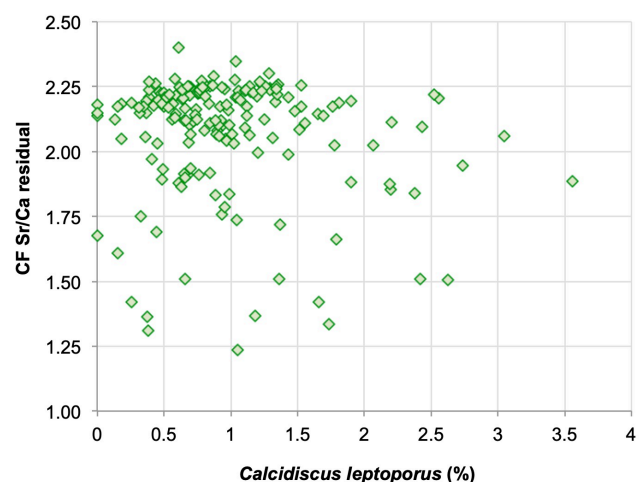


Figure 2

Figures 1 and 2 showing cross-plots of *Calcidiscus leptoporus* with coccolithophore productivity proxy (Fig. 1) and the coccolith fraction (CF) Sr/Ca ratio (mmol/mol).

Table 1

Pearson's Correlation coef.	R	p-value	H0 (5%)
CF Sr/Ca ratio (mmol/mol) vs <i>C. leptoporus</i> (%)	-0.14	0.07	accepted
Coccolithophore productivity proxy vs <i>C. leptoporus</i> (%)	-0.09	0.21	accepted

Line 184 – geophyrocapsids's – lower case I believe.

Yes, we thank the reviewer for this comment and have changed all terms accordingly.

Lines 255- bias from other carbonate phases – from Figure 5 it looks like the trend starts before the cut-off used, from more like 30 mmol/mol Mg/Ca. And could you please clarify which “cut off” you are using, whether this is Sr/Ca below 1.8 mmol/mol or higher values of Mg/Ca? If tracing contamination from other carbonates, would it make more sense to use the Mg/Ca values for the cut-off? For example you could cut off more stringently, at \sim 30 mmol/mol Mg/Ca and yet maintain what look like more robust / primary signals of lower Sr/Ca within data that would pass this criteria. You would lose a few more data points in total, but I think this would be a more defensible cut-off point and rationale.

We thank the reviewer for this comment and have used values of Mg/Ca >30 mmol/mol and agree that this way we have a more defensible cut-off point.

Line – 272 – “This interval represents 39% of the whole sampling variation. . .”, doesn't make sense to me, please clarify. Do you mean something like the clipped data represents 39% of the dynamic range in Sr/Ca of the full sample set?

We thank the reviewer for this comment and we have substituted that sentence with the following one: “The coccolith fraction Sr/Ca ratio results varied between 1.2 and 2.4 mmol/mol (sampling range of 1.16 mmol/mol) with 85 % of the samples falling between 1.8 and 2.3 mmol/mol.”

This means that only 15 % of the samples fall on 70% of the sampling range. And, most of them coincide with the samples with higher bias likelihood from other carbonate phases.

Lines 288-289: is significance level of 80% really enough to be confident that the 6ka peak is real? I'm not really convinced. I don't think this spectral analysis reveals anything and is a distraction for the reader - you have a nice tight coupling to well-resolved climate records (Uk37) and a good age model for making 104-year correlations to N. Atlantic climate records, so I don't think there's a need to try to resolve periodicities independently within this record. I would leave this analysis out.

We agree that we should focus the purpose of this study in long-term processes and not so much on shorter scale changes. Hence, we agree that the spectral analysis might deviate the reader from the most important aspects of our research. Therefore, the spectral analysis was deleted from the revised manuscript.

Lines 355 – 350: are all these paragraphs part of one argument? If so combine.

We thank the reviewer for this comment and have taken it in mind while re-structuring the discussion.

Line 356: Si/Ca fraction. With the preparation methods and uptake by acetic acid digestion, can the authors please clarify which phase / sedimentary component they think the Si is coming from? Is it likely dominated by biogenic Si?

We thank the reviewer for this comment and decided to delete this assumption.

Lines 360 – 365: I think I get what you're trying to say, but this could be expressed more clearly.

We thank the reviewer for this comment and these lines were deleted as a consequence of the previous comment.

Lines 435 – 437 what has MIS12a got to do with MIS 2 and 6? Not clear what your point is here.

We thank the reviewer for this comment and highlight that just for the sake of comparison, we have mentioned other past glacial periods. However, given the re-structuring of the discussion, we expect that the context now is provided in a clearer way in line 450.

DISCUSSION:

In general, the discussion feels long and could have more focus. It feels like you are discussing every aspect of the record from a descriptive perspective, rather than focusing on what the data tell you about processes. I've got specific comments below, but I would focus on the broader longer-term trends and behaviour of the coccolithophore productivity and export system during times of distinct oceanographic conditions (i.e. between the stages) rather than the millennial scale lead and lags (dubious as to how robust these are). It would be great to use these different intervals to try frame clearly articulated conceptual models about how and why growth rate is *coupled* or *decoupled* from organic and inorganic carbon fluxes at different stages. Such conceptual models would have the potential to genuinely inform the thinking of biogeochemical modellers by providing clear patterns of change that should be reproducible by numerical biogeochemical models of these systems. But I think you need to clearly formulate these, in words and ideally schematics, in order for them to take notice of your data. This is also where the multi-proxy approach you take is a clear ADVANTAGE, it's not about "mismatches" in the data, it's about using multi-proxy data to represent the responses of different components of the primary production to export system.

We greatly appreciate these very useful comments from the reviewer and believe that the revised manuscript addresses all of these points.

Section 5.2 – looking at the records, I think it's a matter of scale at which they are interrogated. Yes, they are subtly different, but they also preserve some of the same features with reasonable fidelity – for example there are broad trends from MIS 12 to 11, to 10, to 9 that are conserved between the proxies. I would consider taking off the "Mismatch" from your title to this section, to give you space to consider both the agreements and the divergence. This would be more helpful in the communication of the key findings of the study – point out the agreements first and then suggest the mismatches.

We appreciate this comment and we have taken it into account while re-structuring the discussion. We have divided the discussion into two sections: the first discussed the surface ocean processes that have determined past coccolithophore productivity, and a second one where we have followed the reviewer's suggestions and analysed the coccolithophore productivity (surface signal) with the nannofossil and alkenone accumulation rates (that combine both surface and bottom signals) as linked components of the marine carbon cycle.

Following on from this Line 474 – alkenone and n-alkane fluxes mostly reflect conditions of increased export / preservation of organics. Maybe, but I'm not sure that this can, or should, then be decoupled from "rather than coccolithophore growth". . . in line 475. The first order coupling with some features of the coccolith accumulation rates (NAR) and the alkenones, and also your Sr/Ca measure – e.g. the transition from MIS 12 to 11 – would suggest a coupled system change, and this could be the case if increase coccolithophore production was part of the driver of increased general MAR and organic export and accumulation rates? I just wouldn't be so quick to decouple these components.

We appreciate this comment and we have taken it into account while re-structuring the discussion. The second section of the revised manuscript mentions the ballasting effect of calcite (line 475) and we build our argumentation differently from the original version of the manuscript (please read the argumentation given in lines 485-503. In the last lines of this paragraph we now conclude that both our record and the alkenone accumulation rate evidence indeed increased coccolithophore productivity.

Lines 477 – 485: leads and lags of the alkenone versus Sr/Ca records of <1000 ka – is this getting down to the resolution of sampling uncertainty / offset? Were the records based on the same sample set?

We appreciate this reviewer's comment. As previously mentioned, we deleted the spectral analyses and related text from the revised manuscript.

Figure 8 – useful to have the Uk37 temperatures on this figure as well for reference.

This has been changed accordingly; we agree that it offers a better analysis of the records.

Line 498 – “the NAR reveals large amplitude shifts during times of high coccolithophore productivity” – this seems to be missing the point. The Sr/Ca also shows large amplitude shifts within this period, arguably larger than the NAR. The point is that the NAR steps down substantially from MIS 11b to 11a, whereas Sr/Ca remains high (as you say in preceding sentence). This to me is the interesting system change, and there's a reverse trend in alkenone MAR (gentle rise in values into MIS 11a). Could this be a seasonality thing between Sr/Ca and NAR? I.e. growing faster (higher Sr/Ca) but for a shorter growth season (less coccoliths)? Seasonality could (maybe?) also be coupled with more efficient export and preservation of organics (alkenone MARs)? Ah, yes you come to this in Section 5.2.3. But, (see comment below), I think you could condense, simplify and make your interpretation of the key points more clearly in the discussion; including integrating 5.2.3. with these discussions of the data.

Similarly to the previous reviewer's comment on the coupling/decoupling of our coccolithophore productivity and alkenone data, we also analysed the coccolithophore productivity reconstruction and the nannofossil accumulation rate accordingly. We explain this further on the paragraph starting in line 538 of the revised manuscript.

Lines 505 – 509: again the question would be about sampling uncertainty between the records – can you confirm that these leads / lags are meaningful on the sub-millennial scale? I'm just pushing back, because, as a reader, they do not convey a strong argument about process or feature of the data that I would be confident in. This feeds into a general point about the discussion – I think this could be edited down quite considerably, so that your key points are more clearly and forcefully stated (and more easily digested by the reader!). This cross-spectral analysis doesn't add anything for me.

We greatly appreciated the reviewer for his comments and we have deleted the spectral analysis.

-Reviewer 2

The paper by Catarina Cavaleiro and collaborators entitled ‘Coccolithophore productivity at the western Iberian Margin during the middle Pleistocene (310 – 455 ka) – evidence from coccolith Sr/Ca data’ examines the geochemical response (coccolith Sr/Ca elemental data) across the MIS12 – MIS9 time slice offshore Portugal. Based on published coccolithophorid culture finding, the Authors use the abundance of strontium relative to calcite in fossil coccoliths measure by ICP-AES to derive a palaeoproductivity index during the rapid climatic oscillations of the Pleistocene. The region of interest typified by the Portugal Current System was previously documented in terms of changes in the currentology, sea surface temperatures (among other key climate-sensitive data) in a bunch of publications (cited in the paper). The authors used this well-established framework to interpret fluctuations in Sr/Ca ratios and productivity in the sunlit waters. They also discuss their data at the level of the phytoplanktonic ecosystem as they argue that coccolithophorid growth (and productivity) is dictated by macro and micronutrient availability and the competition with diatoms. They mainly focus their biogeochemical discussion on MIS 12-10 showing higher productivity at the beginning of these climate transitions. Playing at different timescales, they ultimately compare their coccolithophorid productivity indices to the available i) alkenone fluxes and ii) nannofossils accumulation rates in published literature and found some coherencies and discrepancies.

I am generally supportive of publication of this work in *Climate of the Past*. I have, however, a number of comments and questions, which I hope the Authors will find fair and useful to prepare

their revisions.

General comments

- It would be good to state what was measured exactly. ‘Coccolith fraction’ is not sufficient as the less than 20 micron filtrate may contain many non-coccolith particles. Some photos will be welcome from key samples to illustrate this.

We appreciate the reviewer’s comment and we have changed the previous sentence to the following one “To obtain the coccolith fraction (CF) Sr/Ca record, ~250 mg of freeze-dried sample was collected and suspended in 2% ammonia (to avoid carbonate dissolution) and sieved through a 20 μm mesh. This sieving aimed to separate the coccoliths contained in the so-called coccolith fraction (<20 μm) from mostly foraminifera and their fragments, and other larger microfossils or sediment components.”

Indeed, the coccolith fraction may contain non-coccolith particles. However, as explained in the methods section, all samples were treated to avoid Sr contamination from non-carbonate particles (assuming that foraminifera and foraminifera fragments were extracted during sieving). Plus, these samples were counted under a light microscope by the researchers Ornella Amore and Eliana Palumbo whose work is already published (Amore et al., 2012; Palumbo et al., 2013) with no mentioning to other non-particles present in the samples (which, by the way, were not sieved prior to the preparation for the observation under the microscope). According to the reviewer’s request, we provide the only existing available picture (see below) from site MD03-2699, sample 1898 (referring to the depth in the core – 1898 cm) and with a corresponding age of 485 kyr (not covered in our research). Note that this photo was taken during a master class exercise and consequently it tried to gather as many different coccoliths in picture as possible.

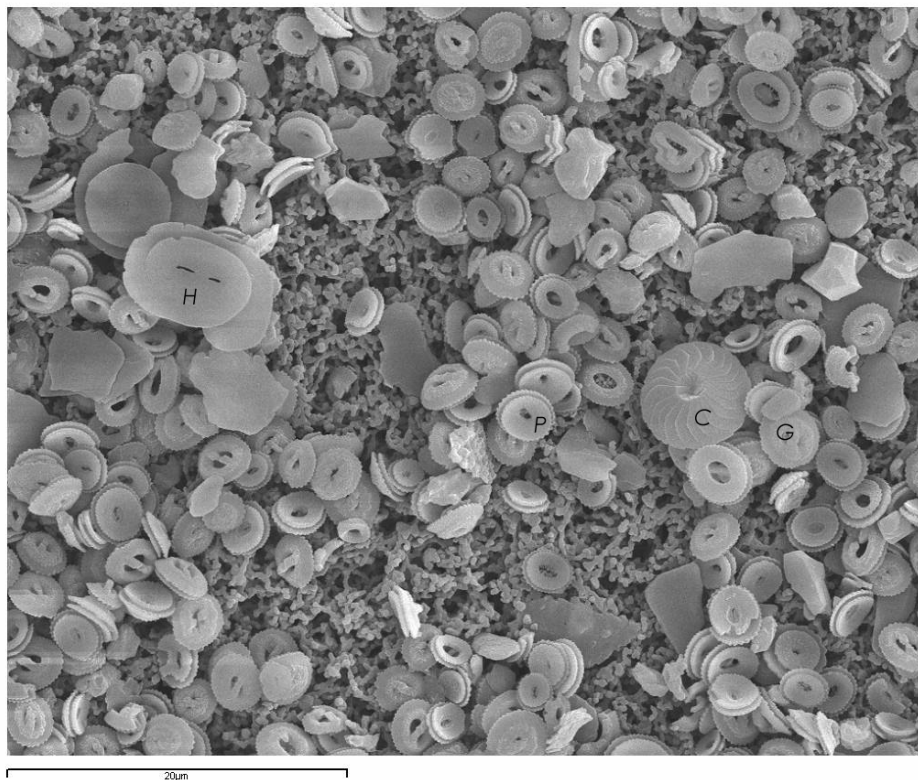


Figure 1 – SEM picture from site MD03-2699, at 1898 cm with a corresponding age of 485 kyr. The irregular surface background is the filter and several coccoliths from different species can be seen, namely Calcidiscus leptoporus, Helicosphaera carteri and gephirocapsids.

- There was this nice paper by Omta et al.) that came out a few years back (On the potential role of marine calcifiers in glacial-interglacial dynamics - doi:10.1002/gbc.20060) in which an elegant model linking ocean alkalinity and the flourishing of coccolithophores at the inception of deglacial periods (with a possible role on the deglaciation). This paper has been omitted in the

present study. I urge the Authors to explore such a control on their productivity data. Even if the periods are not necessarily the same, another useful related paper is that by Duchamp-Alphonse developing the carbonate counter-pump aspects (Enhanced ocean-atmosphere carbon partitioning via the carbonate counter pump during the last deglacial – doi:10.1038/s41467-018-04625-7). What I am trying to say is that the Authors did a pretty good job in integrating local and regional data but quantitatively understanding pelagic calcification requires a bigger biogeochemical picture. We greatly appreciate this reviewer's comment and suggested further reading. We have now included these references in the paper and have re-structured the discussion. The second section of the revised manuscript mentions carbon cycle components, such as the ballasting effect of calcite (line 475). Consequently, our argumentation now differs from the original version of the manuscript. Please check the major changes done in the manuscript, affecting all discussion chapters.

- Sentence line 283 'We would like to stress that our study focuses on the qualitative characteristics of the coccolithophore paleoproductivity record, rather than quantitatively estimating the productivity of coccolithophores.' is misleading and made me doubt about my understanding of the paper. If the Authors interpret Sr/Ca ratios, they intrinsically develop a quantitative approach pertaining productivity in the surface waters.

We thank the reviewer's comment and we would like to stress that this sentence was included in the manuscript to clarify the reader that the CF Sr/Ca ratio is not an absolute productivity proxy neither does it allow for the calculation of an absolute value of marine production of organic carbon or calcium carbonate by coccolithophores.

- Removing the temperature effect from Sr/Ca data to derive productivity component only. I am still debating with myself to be honest. When I read the paper for the first time, I found that it was a good idea. But the more I think, the more I believe that this is not. Both calcification rates and temperatures (and the control of the latter on the former) synergistically dictate Sr/Ca coccolith ratios. Thus dissecting the proxy may induce an artificial bias. I leave these thoughts to the Authors for their revisions...

- Emerging from the previous point, the heart of the Sr/Ca productivity proxy is poorly approached in this paper. The Authors mix cellular growth rate, primary productivity, and calcification rates. This is only calcification rates that control the substitution of Sr to Ca. Yet, culture data are unable to properly measure calcification rates, as they only document the bulk over the course of the batch experiments (See the Appendices in Stoll et al. 'Climate proxies from Sr/Ca of coccolith calcite: Calibrations from continuous culture of *Emiliania huxleyi*' published in 2002 in GCA). Thus, the generalisation of the proxy to productivity is far-fetched, as it implicitly means primary productivity in turn leading to the strength of the biological pump. I think that the Authors should clarify this.

We thank the reviewer's comment and we will further clarify these points in the revised manuscript. We further acknowledge the extent work already done in correlating coccolith Sr/Ca ratio with coccolithophore productivity. Stoll and Schrag (2000) initially suggested that the CF Sr/Ca ratios are strongly controlled by coccolithophorid growth and calcification rate. Stoll et al. (2002a; Potential and limitations of proxy) and Stoll et al. (2002b; *E. huxleyi* cultures), Stoll et al. (2002c; multi species cultures), Stoll et al. (2007a; Arabian and Sargassum seas) and Stoll et al. (2007b; Bay of Bengal) used culture records, sediment traps and sediment samples to confirm the relationship between coccolith Sr/Ca ratios and coccolithophore productivity (coccolithophore growth rate and coccosphere export). Furthermore, Stoll et al. (2002a) and Mejia et al. (2013) clearly stated that the temperature effect on the CF Sr/Ca must be addressed when reconstructing past coccolithophore productivity. Indeed, in our current study the extraction of the temperature effect does not represent a major change of the original curve. However, Cavaleiro et al. (2018) show a final coccolithophore productivity record notably different from the original coccolith fraction Sr/Ca curve due to the large influence of temperature in that area. Our temperature correction in the Iberian margin site reinforces that, contrary to the open mid-latitudinal North Atlantic, the temperature changes in the

Iberian margin do not seem to have affected the coccolith fraction Sr/Ca and consequently coccolithophore productivity. In addition, the possibility to use a proxy that is independent of accumulation rates allows comparison with commonly used “coccolithophore productivity proxies” such as nannofossil accumulation ratios and alkenone export from which coccolithophore productivity, in the ancient photic layer, is commonly inferred from. Finally, the term productivity is thus in this research used as a coccolithophore productivity proxy directly associated with coccolith calcification rate and generally associated with increased cell division and growth of coccolithophores that could lead to increased particulate organic matter and calcium carbonate export.

- The Authors spent considerable effort (and space in the manuscript) to try and find a good match between their coccolithophore productivity and the sedimentation of Point 1 coccolith-derived calcite (NAR) on one hand, and Point 2 coccolith-derived compound-specific organic matter (alkenones) on the other hand. Point 1 For the reasons outlined above, the Sr/Ca has not to scale with the bulk production (-ity) of calcite. This geochemical proxy has to do with intracellular processes why the production of calcite is also related ecologically with the density of cells in seawater and cellular division rates. Point 2 We know that alkenones are not only synthesized by the coccolithophores but also by other non-calcifying haptophytes (incl. naked coccolithophores). Furthermore, the export of calcite and organic matter from the top of the water column down to the seafloor obey to different processes (as their on the seafloor and during sedimentary burial diagenesis).

Therefore, I cannot see why all these parameters should scale. I am not aware of any sedimentary succession in which this is the case. I am happy to be wrong though.

We appreciate this reviewer's comment and believe that the re-structuring of the paper, namely of the second section, these point are addressed. The second section of the revised manuscript mentions carbon cycle components, such as the ballasting effect of calcite (line 475), and suggests conceptual models for the carbon cycle and its different components. Please check the major changes the manuscript suffered in the Discussion.

- I personally disagree with the fact the Si and Fe concentrations relative to Ca are meaningful in such a sedimentary study nor that they reflect the palaeoconcentrations of these elements. Si and Fe are very tricky to measure and it is unlikely that the measurements reflect the composition of coccolith calcite. Even if it was the case, by which means (proxy) the coccolith Si/Ca ratios would reflect the concentration of silicic acid in ambient waters?

We thank the reviewer's comment and we have decided to delete these assumptions from the manuscript.

- The Authors have managed to lose me with the concept of phenology they are trying to introduce. This is a black box concept and this is very misleading or at least not clear at all. Could they elaborate?

We thank the reviewer for this comment and we have decided to delete the coccolithophore phenology change discussion, which was partially linked to the spectral analyses results that have now been omitted from the manuscript. Coccolithophores' phenology (meaning the yearly timing at which they had higher productivity) most certainly changed throughout glacial-interglacial cycles. However, we have not focused on phenology changes in the paper because we believe it would distract the reader from the main findings.

- I found the statistics very poorly treated in the manuscript.

We thank the reviewer's comment and, where possible and according to what we assumed could be improved, we added more information to clarify for the reader the statistical analysis applied. If the reviewer refers to the spectral analysis presented in the previous version of the manuscript, we would like to clarify that, also based on reviewer 1 comment's, the spectral analysis was deleted

from the latest version of the manuscript because it distracted the reader from the most important aspects of this research.

Specific comments

Pg 1 Line 16. Perhaps use Carbonate Counter-Pump instead?

We thank the reviewer for this comment and we have changed the term accordingly.

Pg 1 Line 30. This what?

We thank the reviewer's comment and since the abstract was changed, we believe this issue was addressed.

Pg 1 Line 33. Not clear to me.

We thank the reviewer's comment and this is addressed in the body of the paper to clarify why changes in climate could influence changes in the coccolithophore phenology.

Pg 2 Line 40. Circumvoluted sentence. Consider splitting it.

We agree with the reviewer and since the introduction was also re-structured and re-written, we believe this issue has been addressed.

Pg 2 Line 59. I disagree with this statement (see General points).

We thank the reviewer's comment and following the answer already given to the General points, we only add that we have added references supporting that calcification rate is a function of growth rate in coccolithophores, namely Balch et al. (1996) and Daniels et al. (2018).

Pg 3 Line 68. I wonder whether the changes in size of geophyrocapsid coccoliths could influence the Sr/Ca ratio

We thank the reviewer's comment but believe this is out of the scope of our study since it is not our intention to better understand how coccolith Sr/Ca varies with the size of geophyrocapsa coccoliths.

Pg 4 Line 97. Poorly defined in terms of what?

We thank the reviewer's comment but believe this is out of the scope of our research since it is not our intention to better describe the Portugal current system. We can however substitute "poorly defined due to" by "with" for simplification.

Pg 5 Line 122. Minimum numbers. Do you mean absolute or relative abundances?

We thank the reviewer's comment and we will clarify the text by stating "absolute abundances" instead of "numbers", now in line 146.

Pg 6 Line 156. The less than 20 micron fraction contain non coccolith particles. The Authors should do a better job in the characterization of the calcite / dolomite particles analysed. This is crucial.

We thank the reviewer's comment and believe we have already replied to this issue above, under the "general points" comments.

Section 3.4. I don't understand what is the relevance of this.

We agree that this spectral analysis might deviate the reader from the most important aspects of our research and for that reason we have deleted it from the revised version.

Section 4.1. belongs to the discussion. Section 4.2 should come first noting that the description of the results is extremely skinny.

We thank the reviewer for this comment but disagree. In our opinion, sections 4.1 and 4.2 should remain in the results section. We agree that some discussion of the results is done here, but the discussion mostly serves to validate our coccolithophore productivity reconstruction in order for it

to be discussed further in the discussion chapters, namely the reasons why coccolithophore productivity changed and why coupling/decoupling with other coccolithophore proxies might have occurred.

Figure 4. Please make the ages more legible.

We thank the reviewer's comment and the figure has been changed accordingly, as well as in figure 3.

Figure 5. What is the significance of the anti-correlation between Mg and Sr?

We thank the reviewer's comment and this information was added to the manuscript. The p-value of the Pearson correlation is 0, therefore the relationship between the coccolith fraction and the coccolithophore productivity and the coccolith fraction Mg/Ca is highly significant, as expected. It is the outliers, or higher values of Mg/Ca that are associated with very low CF Sr/Ca ratios and coccolithophore productivity results, mostly associated to abrupt and cold millennial-scale events.

Figure 6 is unnecessary in my opinion.

We agree that this spectral analysis might deviate the reader from the most important aspects of our research. Therefore, this analysis was left out of the revised manuscript.

Pg 14 Line 303. See my general comment on temperature and productivity on Sr/Ca ratios.

We thank the reviewer's comment and believe we have already replied to this issue above, under the "general points" comments.

Pg 16 Line 333. What do you refer to with 'opportunistic and fast growing species' here?

We appreciate the reviewer's comment and clarify that by "opportunistic and fast growing species" we refer to species with r-selected strategies, meaning that they react more rapidly with higher growth rates than k-selected species that are adapted to low nutrients and have lower growth rates (an example of a paper focusing on coccolithophore dynamic in the Iberian margin also refer these more biological terms: Guerreiro, C., Oliveira, A., De Stigter, H., Cachão, M., Sá, C., Borges, C., Cros, L., Santos, A., Fortuño, J. M. and Rodrigues, A.: Late winter coccolithophore bloom off central Portugal in response to river discharge and upwelling, *Cont. Shelf Res.*, 59, 65–83, doi:10.1016/j.csr.2013.04.016, 2013.)

Pg 16 Line 355. Methodologically unjustified even using 'weak' acid.

We thank the reviewer for this comment and we have decided to delete this assumption that higher coccolith fraction Si/Ca and Fe/Ca could evidence higher competition with diatoms.

Pg 17 Line 361. Sentence not clear and too long.

We thank the reviewer for this comment and we have decided to delete this assumption that higher coccolith fraction Si/Ca and Fe/Ca could evidence higher competition with diatoms.

Pg 17 Line 371. Decrease of the SST.

We thank the reviewer's comment and it has been changed accordingly.

Pg 18 Line 421. I am not following the logic here. Are the Authors trying to say that the ice coverage reached the studied area?

We thank the reviewer's comment and clarify that we have not stated or suggested that ice coverage reached the Iberian margin. We are stating research that has found evidences of the presence of melting icebergs in the eastern mid-latitudinal North Atlantic and in western Iberian margin during rapid millennial-scale events, namely Alonso-Garcia et al. (2011), Hodell et al. (2008), McManus et al. (1999), Rodrigues et al. (2011, 2017). We believe that it is out of the scope of this study to infer sea-ice coverage in the Iberian margin during the analyzed interval.

Pg 19 Line 449. I don't understand the point that the Authors are trying to make here.

We thank the reviewer for this comment and we have decided to delete this assumption that higher coccolith fraction Si/Ca and Fe/Ca could evidence higher competition with diatoms.

Pg 19 Line 454. Visual comparison of what?

We thank the reviewer's comment and further clarify that we refer that the records of coccolithophore productivity, nannofossil accumulation rate and alkenone flux were compared visually.

Pg 19 Line 457. An illustration of the poor statistical approach here. ..

We thank the reviewer for this comment but given the re-structuring of the discussion we no longer present such data.

Pg 21 Lines 477- 492 and figure 9 are not necessary.

We agree that this spectral analysis might deviate the reader from the most important aspects of our research. Therefore, this analysis was left out of the revised manuscript.

Coccolithophore productivity at the western Iberian Margin during the middle Pleistocene (310 – 455 ka) – evidence from coccolith Sr/Ca data

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Abstract. Coccolithophores contribute significantly to marine primary productivity and play a unique role in ocean biogeochemistry by using carbon for photosynthesis (soft tissue pump) and for calcification (carbonate counter pump). Despite the importance of including coccolithophores in Earth System Models to allow better predictions of the climate system's responses to planetary change, the reconstruction of coccolithophore productivity mostly relied on proxies dependent on accumulation and sedimentation rates, and preservation conditions. In this study we used an independent proxy, based on the coccolith fraction (CF) Sr/Ca ratio, to reconstruct coccolithophore productivity. We studied the marine sediment core MD03-2699 from the western Iberian margin (IbM), concentrating on glacial/interglacial cycles of Marine Isotopic Stage (MIS) 12 to MIS 9. We found that IbM coccolithophore productivity was controlled by changes in the oceanographic conditions, such as in sea surface temperature (SST) and nutrient availability, and by competition with other phytoplankton groups. Long-term coccolithophore productivity was primarily affected by variations in the dominant surface water mass. Polar and subpolar surface waters during glacial substages were associated with decreased coccolithophore productivity, with strongest productivity minima concomitant with Heinrich-type events (HtE). Subtropical, nutrient-poorer waters, increased terrigenous input and moderate to strong upwelling during the deglaciation and early MIS11 are hypothesized to have attributed a competitive advantage to diatoms in detriment of coccolithophores, resulting in intermediate coccolithophore productivity levels. During the progression towards full glacial conditions increasing presence of nutrient-rich waters, related to growing influence of transitional surface waters and/or intensified upwelling, probably stimulated coccolithophore productivity to maxima following the rapid depletion of silica by diatoms. We present conceptual models of the carbon and carbonate cycle components for the IbM in different time slices that might serve as a basis for further investigation and modeling experiments.

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1 Introduction

Coccolithophores play a unique role in ocean biogeochemistry using carbon for both photosynthesis and calcification (e.g. Rost and Riebesell, 2004; Westbroek et al., 1993), and contributing up to 60 % to the total calcium carbonate in the ocean (Flores and Sierro, 2007). During the Mid Brunhes interval of Marine Isotope Stage (MIS) 14 to MIS 9, when the assemblages were vastly dominated by gephyrocapsids (Baumann and Freitag, 2004; Saavedra-Pellitero et al., 2017), coccolithophores provided >80 % of the total oceanic calcium carbonate. And, several studies hypothesized a relevant role of coccolithophores on glacial-interglacial dynamics (Duchamp-Alphonse et al., 2018; McClelland et al., 2016; Omta et al., 2013; Rickaby et al., 2007; Saavedra-Pellitero et al., 2017) but there are uncertainties arising from the complex interactions between coccolithophore productivity and growth rates, nutrient dynamics and competition with non-calcifiers, seasonality and export of both carbonate and organic carbon and the final accumulation/burial rates (Balch, 2018; Duchamp-Alphonse et al., 2018; McClelland et al., 2016; Omta et al., 2013; Rickaby et al., 2007; Ridgwell and Zeebe, 2005; Saavedra-Pellitero et al., 2017). These uncertainties make the inclusion of biogeochemical processes related to coccolithophores in Earth System models challenging. In order to provide useful information to modellers, it is thus fundamental to test the dynamic response of the various components of the system over different climatic scenarios through the use of multiproxy studies involving the reconstruction of coccolithophores growth rates and export of both their organic and inorganic compounds, namely alkenones and nannofossil accumulation rate (AlkAR and NAR, respectively). Since coccolithophores are sensitive to rapid fluctuations in temperature, salinity, nutrients, and turbidity of surface waters (Baumann et al., 2005; McIntyre and Bé, 1967), their calcareous remains, the coccoliths, retrieved from deep-sea sediments have been used extensively to reconstruct paleoenvironmental conditions (Amore et al., 2012; Baumann et al., 2005; Beaufort et al., 2001; Flores et al., 1997; Maiorano et al., 2015; Marino et al., 2014; McIntyre and Molino, 1996; Saavedra-Pellitero et al., 2017). However, coccolithophore paleoproductivity reconstruction has been tentative and mostly relied on proxies dependent not only on the extent of the supply but also on dilution by mineral matter, changes in sedimentation or accumulation rates, as well as preservation conditions (Rullkötter, 2006). Beaufort et al. (1997) proposed a proxy to quantitatively reconstruct coccolithophore productivity, but its applicability is unfortunately limited to latitudes between 30°N and 30°S (Hernández-Almeida et al., 2019). A widely used alternative proxy is the coccolith fraction Sr/Ca (CF Sr/Ca) ratio that is independent of accumulation rate (Cavaleiro et al., 2018; Mejía et al., 2014; Saavedra-Pellitero et al., 2017; Tangunan et al., 2017). Coccolithophores construct coccoliths internally within their cell and several studies show the direct and proportional relationship between the Sr/Ca ratio of the coccolith and the coccolith calcification rate. Calcification rate is a function of growth rate (e.g. Daniels et al., 2018) and therefore of coccolithophore productivity (Balch et al., 1996; Rickaby et al., 2007; Stoll and Schrag, 2000). The faster coccolithophores grow, the faster they calcify and more Sr is incorporated into the calcite lattice of their coccoliths (Stoll et al., 2002b, 2002a; Stoll and Schrag, 2000). With temperature and assemblage effects considered (see Material and methods section), the SST corrected CF Sr/Ca curve (or residual curve)

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can be expected to reflect coccolithophores' growth rate and thus their productivity qualitatively (Müller et al., 2014; Stoll et al., 2002a).

Coccolithophores research has been gaining increased attention because of their potential relevant role on the carbon cycle (Ridgwell and Zeebe, 2005). The carbon cycle underwent major changes during the past 800 kyr as evidenced for example by the glacial-interglacial variations in atmospheric carbon dioxide (atmCO₂) concentrations (Lüthi et al., 2008). Though the changing orbital parameters of Earth (i.e. the Milankovitch cycles) are undoubtedly related to the glacial-interglacial cycles (e.g. Berger, 1988), the mechanisms driving the almost 80 ppmv lower atmCO₂ concentrations during glacial periods and the rapid rise during deglaciations are yet to be described and modelled accurately (Barker et al., 2006; Duchamp-Alphonse et al., 2018; Omta et al., 2013; Ridgwell and Zeebe, 2005). Since the ocean contains 60 times more carbon than the atmosphere, modest changes in its holding capacity could largely affect atmCO₂ (e.g. Sarmiento and Gruber, 2007). It is clear that atmCO₂ changes are a consequence of several factors interplaying and not a single mechanism at work (Archer et al., 2000; Sigman and Boyle, 2000). Yet, theories such as the "coral reef hypothesis" (Berger, 1982), "carbonate compensation" (Broecker and Peng, 1987) and "rain ratio hypothesis" (Archer and Maier-Reimer, 1994) give the marine carbonate cycle a determinant role on atmCO₂ changes on time scales shorter than 100 kyr (e.g. Ridgwell and Zeebe, 2005; Broecker, 1982).

Our studied period falls into the mid-Brunhes interval (Barker et al., 2006; Baumann and Freitag, 2004; Jansen et al., 1986), when, starting with MIS 11c, interglacial atmCO₂ levels were comparable to the Holocene (Lüthi et al., 2008). The Mid Brunhes interval also has the advantage to cover the *Gephyrocapsa caribbeanica* acme (Baumann and Freitag, 2004), reducing the likelihood of biasing effect of the CF Sr/Ca ratio from changing coccolith assemblages (see Material and methods section). It encompasses different climatic scenarios, from late MIS 12 to early MIS 9, straddling the harsh glacial of MIS 12 and the prolonged MIS 11c interglacial period, considered one of the best analogues for the current MIS 1 interglacial (Berger et al., 2015; Candy et al., 2014; Loutre and Berger, 2003; Oliveira et al., 2018). The deglaciation and termination (T) V is considered one of the longest deglaciations (Tzedakis et al., 2012; Vázquez Riveiros et al., 2013), and the highest glacial-interglacial amplitude change of the last one million years (Lisiecki and Raymo, 2005), allowing a more detailed analysis due to the high sedimentation rates that characterize the western Iberian Margin (IbM).

The western IbM embodies the northern section of the Canary Current Eastern Boundary Upwelling System (Aristegui et al., 2009; Mason et al., 2006). The seasonal meridional shifts of the atmospheric highs revert the upwelling favouring equatorward winds seasonally to become poleward in autumn and winter (Mason et al., 2006). Despite representing only 10 % of the ocean, the Eastern Boundary Upwelling Systems contribute 80 to 90 % to the oceanic new production (Hill et al., 1998; Ryther, 1969). These systems are sensitive to climate change by responding to shifts in processes that regulate ocean stratification, upwelling, wind stress, dust supply, and basin-wide circulation (Freon et al., 2009). However, to infer the role of climate modes in modulating air-sea CO₂ fluxes in such a complex environment is challenging although this knowledge is crucial to improve our ability to predict future evolution of atmCO₂ (Brady et al., 2019).

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In this study we reconstruct coccolithophore productivity from the CF Sr/Ca ratio record at Site MD03-2699, an area under the influence of the Canary Current upwelling system, and particularly sensitive to global climate change (Hodell et al., 2015; Martrat et al., 2007; Oliveira et al., 2016; Rodrigues et al., 2017) and thus optimal to evaluate this phytoplankton group's behaviour to varying conditions. Using a multi-proxy approach, we compare the CF Sr/Ca data with the total alkenones concentration (i.e., the exported organic carbon produced by coccolithophores), as well as with the nannofossil accumulation rates (NAR) in order to investigate the processes of production versus export and preservation, of both organic and inorganic carbon. This approach allows us to gain a better understanding of (1) the coupling/decoupling between the different proxies used to reconstruct coccolithophores' productivity and (2) the role of upwelling strength and terrigenous input in nutrient competition under different climate conditions, such as glacial, interglacial, deglaciations and the transition from interglacial to glacial conditions.

2 Regional setting

2.1 Present hydrography of the IbM

The study area is located at the north-eastern edge of the subtropical gyre and influenced by a southward flowing branch of the North Atlantic Current forming the Portugal Current system (Fig. 1). The Portugal Current system is mainly characterized by a slow, southerly flow with intricate interaction between coastal and offshore currents (Peliz et al., 2005; Relvas et al., 2007), bottom topography and water mass convergence (Bischof et al., 2003). One of the most important features of the IbM is the surface circulation's seasonality as a response to the combined position of the Azores high and the Icelandic low-pressure systems (Barton, 2001; Haynes and Barton, 2018; Relvas et al., 2007). During summer, the migration of the Azores high to the central Atlantic exposes the IbM to northerly Trade Winds, strengthening the Portugal Current and forcing the upper layer (150 to 200 m) to flow towards the equator. The induced offshore Ekman transport allows colder, less salty and nutrient-rich subsurface water to rise to the surface (Alvarez et al., 2011; Fiúza, 1983) leads to high primary production (Figueiras et al., 2002; Fraga, 1981; Tenore et al., 2018). The upwelled water is the Eastern North Atlantic Central Water (ENACW) that has two different origins: a less saline, colder, nutrient richer water mass of subpolar origin (ENACWsp) formed along with the Subpolar Mode Water in the Rockall Plateau region, and a warmer, saltier, nutrient poorer subtropical branch (ENACWst) formed along the Azores Front (Fiúza et al., 1984). Depending on the wind strength intensity either type can be upwelled (Fiúza et al., 1984), and in the current interglacial ENACWsp is mostly upwelled in the northern western coast of the Iberian peninsula, while ENACWst is mostly upwelled in the southern area of the western Iberian peninsula (Mason et al., 2006). The upwelling season typically lasts from late spring into summer (May to September) and upwelling filaments are mostly observed off the most prominent capes (Fiúza et al., 1982). Upwelling plumes and other mesoscale features can spread zonally offshore to distances of 200 km or more (Sousa and Bricaud, 1992). In autumn and winter the seasonal meridional shifts of the atmospheric highs revert the upwelling favouring equatorward winds to become poleward (Mason et al., 2006). The Azores high located further south and the Icelandic low intensification,

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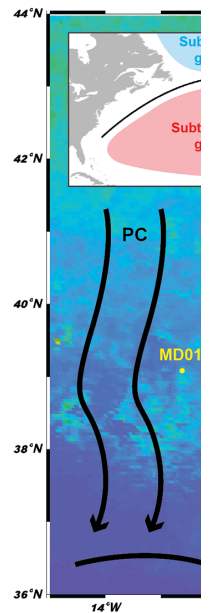
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forces the wind regime to become more southerly, enabling the near shore Iberian Poleward Current, which brings warmer and more oligotrophic waters from the Azores Current to the shelf and upper slope areas along the western IbM (Fiúza, 1983; Peliz et al., 2005; Vitorino et al., 2002). Downwelling can occur during this period but winter-time fresh-water discharges from the major rivers are also a relevant feature, namely from the Tagus river that crosses the Iberian Peninsula, carrying a major load of suspended organic-rich sediments onto the continental shelf off Lisbon (Cabeçadas and Brogueira, 1998) and the Estremadura spur where our sediment core is located.

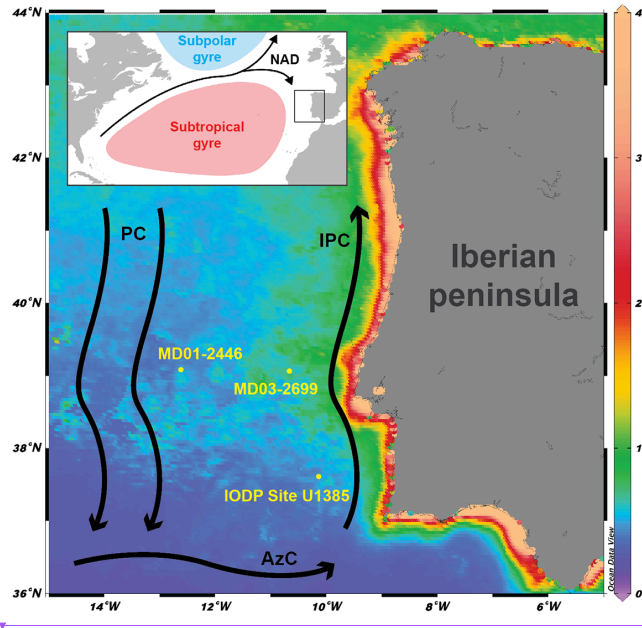


Figure 1 Core location and major currents in the area: NAD = North Atlantic Drift; PC = Portugal Current; IPC = Iberian Poleward Current; AzC = Azores Current. Background: chlorophyll *a* concentration (mg.m^{-3} ; March, April and May average, 2003-2018) derived from MODISA satellite data available at <http://disc.sci.gsfc.nasa.gov/giovanni>.

2.2 Present coccolithophore productivity patterns in the IbM

Several studies on coccolithophore abundance and distribution (e.g. Abrantes and Moita, 1999; Cachão et al., 2000; Guerreiro et al., 2013; Moita, 2001) have been performed along the IbM but only two were able to assess annual and seasonal productive cycles (Ausín et al., 2018; Silva, 2008; Silva et al., 2009). Maximum coccolithophores were found during spring and summer, and associated with high irradiance levels (Ausín et al., 2017) and the convergence of warmer,

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more oligotrophic waters with relaxation of northerly winds (Silva, 2008; Silva et al., 2009). Minimum **absolute abundances**, on the other hand, were observed during winter. The competitive dynamics between coccolithophores and diatoms are still not completely understood (Cermeno et al., 2011). Yet, these two main phytoplankton groups dominate the carbon dioxide mediation between the atmosphere and the ocean making them important contributors to the global carbon cycle. Diatoms are known to require more silica (Si) and iron (Fe) than coccolithophores (e.g., Merico et al., 2004) and Tyrrell and Young (2009) suggested that coccolithophore blooms might be associated with Si and Fe surface water depletion. Cermeno et al. (2011) suggested that coccolithophores could outcompete diatoms under steady-state nitrate limitation but under dynamical conditions (introducing nitrate pulses) diatoms outcompeted coccolithophores. They further found that the more frequent the pulses, the more rapidly diatoms outcompeted coccolithophores. The coastal upwelling conditions resemble these dynamical conditions, when pulses of nutrients are brought to the ocean surface. This could explain why Abrantes and Moita (1999) found a phytoplankton dominance of diatoms during upwelling events and a clear dominance of coccolithophores (~ 90 %) during non-upwelling events. Also, Ausin et al. (2017) describe decreasing abundance of diatoms with increasing distance to shore whereas coccolithophores showed higher abundances offshore than in coastal areas. This distribution pattern was maintained during the upwelling season (May to September), when coccolithophores are outcompeted by diatoms, in particular during the longer lasting and more intense upwelling events (Moita, 2001; Silva et al., 2008).

Despite the general acceptance of coccolithophores as a single functional **phytoplanktonic** group associated with low turbulence, low nutrient and high irradiance environments, different species show varying life strategies. Several studies describe increased cell densities, mostly due to blooms of *Emiliania huxleyi* and *Gephyrocapsa oceanica* in the IbM (Moita, 2001; Guerreiro et al., 2013; Silva, 2008; Silva et al., 2009), together with other fast-growing opportunistic phytoplankton genera, such as the diatoms *Chaetoceros* s.l., *Thalassiosira* s.l and *Skeletonema* s.l.. This corroborates coccolithophores' role among early succession taxa, or at least some species, capable of rapid growth in a nutrient-rich environment and most likely explains why Abrantes and Moita (1999) found coccolithophores' distribution in recent sediments to reflect their water column distribution during an upwelling situation **and not the winter conditions**.

3 Material and methods

3.1 Sediment sampling and coccolith fraction separation

In this study we used sediments of core MD03-2699 (39°02.20'N, 10°39.63'W, 1895 m water depth), retrieved from a sediment drift, located ca. 100 km offshore, on the Estremadura promontory (Fig. 1). Sediments were collected using a giant CALYPSO piston corer on board the R/V Marion Dufresne II (PICABIA Cruise, 2003). The sedimentary record is mainly composed of hemipelagic silty clays. For our research, we use the age model published by Voelker et al. (2010). For the CF Sr/Ca ratio, a total of 183 samples were analysed. Samples were taken at 4 cm spacing from the ~7 meters long section (from 1190 to 1898 cm core depth) corresponding to MIS 12 to MIS 9 (~ 455 to 310 ka). This resulted in a temporal resolution of ~775 years, although the oldest part has lower resolution due to the lower sedimentation rates (Rodrigues et al.,

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2011). To obtain the [coccolith fraction \(CF\)](#) Sr/Ca record, ~250 mg of freeze-dried sample was collected and suspended in 2% ammonia (to avoid carbonate dissolution) and sieved through a 20 µm mesh. [This sieving aimed to separate the coccoliths contained in the so-called coccolith fraction \(<20 µm\)](#) from mostly foraminifera and their fragments, and other larger microfossils or sediment components. All sieving material was carefully washed with running tap water and rinsed with distilled water in between samples to avoid cross contamination.

3.2 Sample preparation and Sr/Ca analysis

We followed a three step protocol to clean the sediment samples based on Stoll and Ziveri (2002): (1) addition of 15 ml of MNX reagent (75 mg of hydroxylamine hydrochloride, 6 ml of concentrated ammonia and 9 ml of ultrapure water) for a 12 hours reaction in an automatic shaker. This step reduces Fe and Mn oxyhydroxides that scavenge metals from seawater and contain non-carbonate Sr, (2) addition of 2% ammonia to remove any non-carbonate Sr, e.g., from clays, by exchanging cations (Sr_2^+) with the excess of NH_4^+ , (3) three ultrapure water rinses to extract the ammonia. A weak buffered acid (6 g glacial acetic acid, 7 g ammonium acetate in 1 l of Milli-Q water) was then used to dissolve the coccoliths, minimizing the contribution of ions from non-carbonate phases. The samples were left in acid during 12 hours and the obtained solution was centrifuged, extracted and kept in acid-cleaned centrifuge tubes. A first ICP-AES measurement of Ca was performed by diluting 100 µl of the original sample (2 ml) into 2 ml of ultrapure Millipore water. The samples were subsequently diluted to Ca concentrations similar to the standard solutions. Calibration was conducted following the method described by de Villiers et al. (2002) using standards with constant Ca concentrations and different Sr concentrations to provide Sr/Ca ratios ranging from 0.75 to 4 mmol/mol. All measurements were conducted using the ICP-AES (Thermo ICAP DUO 6300) in the Geology Department at the University of Oviedo with reproducibility better than 0.02 mmol/mol. To infer any possible contamination other metals, such as Fe and Mg, were also measured together with Sr.

3.3 Extraction of paleoproductivity record from the CF Sr/Ca ratios

[Culture studies found a temperature dependence of coccolith Sr/Ca with a 0.03 mmol/mol increase per degree Celsius rise \(Müller et al., 2014; Stoll et al., 2002a\). This signal can, however, be removed from the Sr/Ca record using an independent SST reconstruction, so that the component of variation due to growth rate remains as residual \(e.g., Cavaleiro et al., 2018; Mejia et al., 2014; \(e.g. Cavaleiro et al., 2018; Mejia et al., 2014; Saavedra-Pellitero et al., 2017\). The partitioning of Sr into the coccolith calcite varies among species \(with larger and more heavily calcified coccoliths generally having a higher Sr content than smaller and lighter coccoliths \(Fink et al., 2010; Stoll et al., 2007\)\). Although it has been demonstrated that CF Sr/Ca ratios are not primarily controlled by variations in the coccolith assemblage in the modern ocean \(Barker et al., 2006; Stoll and Schrag, 2000\), the species composition, especially regarding the abundance of heavily calcified coccolithophores \(e.g., *Calcidiscus leptoporus*, *Helicosphaera carteri*, *Coccolithus pelagicus*\), should always be assessed. Our study period falls within the *Gephyrocapsa caribbeanica* acme \(Baumann and Freitag, 2004; Bollmann et al., 1998\), described between MIS 14/13 and MIS 8 when this species dominated \(\$\geq 50\%\$ \) the coccolithophore community globally. In core MD03-2699,](#)

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the gephyrocapsids contribute more than 92 % to the flora (on average 97 %; Fig. 2; Amore et al., 2012), what minimizes the bias of the CF Sr/Ca data due to significant coccolith assemblage changes. The sum of the remaining coccoliths, which may contain the larger and Sr-rich coccoliths *Calcidiscus leptoporus*, *Coccolithus pelagicus* and *Helicosphaera carteri*, averaged 2.8 % (maximum 8.1%, standard deviation 1.6%). If we consider the abundance of the larger and Sr-rich coccoliths *Calcidiscus leptoporus*, *Coccolithus pelagicus* and *Helicosphaera carteri* the average and maximum abundances were, respectively: 1 % and 3.6 %, 0.1 % and 0.3 %, and 0.2 and 1.5 %. Their contribution to the total Sr of the coccolith fraction is, therefore, negligible when compared to the gephyrocapsids' contribution.

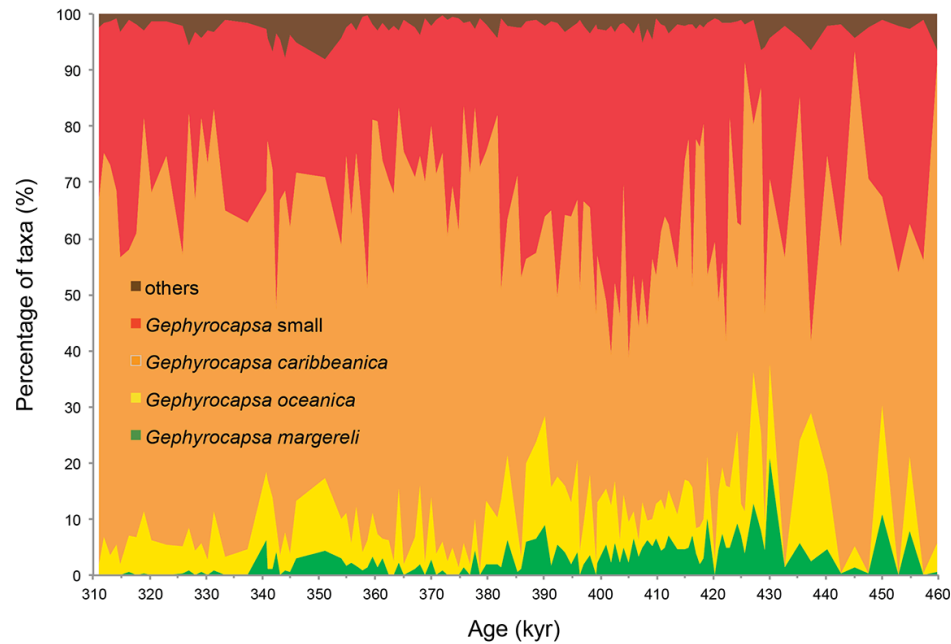


Figure 2 Composition of coccolith assemblages in core MD03-2699 based on Amore et al. (2012). Note that only a minor percentage of coccoliths belong to groups other than the dominant gephyrocapsids. The chronology is based on the age model of Voelker et al. (2010).

With the assemblage changes considered we then moved to the temperature effect extraction. This was extracted from the CF Sr/Ca ratios record following Mejia et al. (2014) and Cavaleiro et al. (2018). We used the multi-species temperature

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dependence to calculate a SST-predicted Sr/Ca curve and the extraction of the temperature effect consisted in subtracting the SST-predicted Sr/Ca values from the initial CF Sr/Ca ratios, Eq. 1:

$$CF \frac{Sr}{Ca} - SST_{predicted} \frac{Sr}{Ca} = CF \frac{Sr}{Ca} residual$$

(1)

405 We calculated the temperature dependence using the $U^{K_{37}}$ -based SST record of core MD03-2699 for the same period (Rodrigues et al., 2011). The alkenones present in the sediments are today mostly produced by a few species of the class Prymnesiophyceae, mainly by the coccolithophores *Emiliania huxleyi* and *Gephyrocapsa oceanica* (Prah1 et al., 1988; Prah1 and Wakeham, 1987), and the $U^{K_{37}}$ -based SST is therefore the best estimator of coccolithophores' habitat temperature. After extraction of the temperature influence, the resultant residual mainly represents coccolithophore calcification and growth rate and consequently coccolithophore long-term productivity fluctuations. We refer to the Sr/Ca residual as coccolithophore productivity because after correcting for the temperature changes, we expect the data to mostly reflect coccolithophore calcification and growth rate and thus their paleoproductivity qualitatively. This coccolithophore productivity record reflects relative productivity change, representing the productivity deviation around the average productivity of the time series.

415 An uncertainty envelope/confidence interval for the CF Sr/Ca residual estimation (shown as a grey envelope in Fig. 3) was calculated using Astrochron (Meyers, 2014) where the upper and lower limits correspond to the Monte Carlo 20 and 80 % confidence interval, respectively. One hundred Monte Carlo simulations were run for each of the 183 data points carrying out propagation of errors accounting for uncertainties measurement of: (1) temperature (with $\sigma = 1.5^{\circ}C$); (2) Sr concentration (with $\sigma = 0.02$ mmol/mol); and (3) the linear regression of temperature versus Sr ($\sigma = 0.12$ mmol/mol).

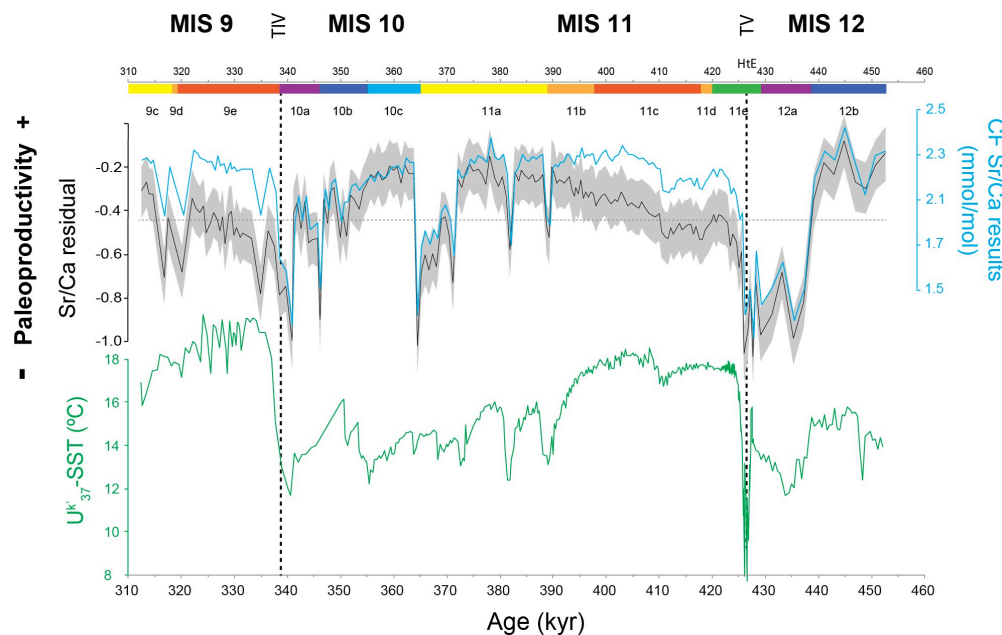
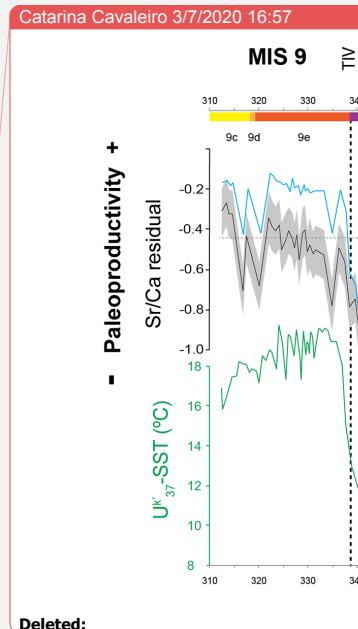


Figure 3 Cocolithophore paleoproductivity results from core MD03-2699: coccolith fraction (CF) Sr/Ca ratio and the resulting CF Sr/Ca residual with confidence interval (grey shading represents Monte Carlo 20-80% interval), U^k_{37} -based reconstruction of sea surface temperature (Rodrigues et al., 2011). The horizontal dashed line marks the average value for CF Sr/Ca and CF Sr/Ca residual. The chronology is based on the age model of Voelker et al. (2010), Marine isotope stages (MIS) and substages marked according to Ralsback et al. (2015), vertical dashed lines highlight Terminations V and IV.

4 Results

4.1 Potential influence of detrital carbonate on the CF Sr/Ca data

When present, detrital carbonate could negatively bias any CF Sr/Ca ratio (and any coccolithophore productivity reconstruction) by increasing the background carbonate in relation to the coccolith carbonate. In our study, we considered potential sources of detrital carbonate biasing the CF Sr/Ca results: (1) ice-rafted debris (IRD) discharged by melting icebergs; (2) aeolian input and (3) riverine discharge. Deposition of carbonate/dolomite rich IRD is linked to extremely cold and lower salinity conditions (Fig. 4), namely Heinrich-type events (HtE; Andrews and Voelker, 2018; Hodell et al., 2008; Rodrigues et al., 2011; Salgueiro et al., 2010; Stein et al., 2009). Ice-rafted material, especially the fine-grained material, also



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contains reworked coccoliths, which can be Tertiary or Cretaceous in age (Marino et al., 2011, 2014; Rahman, 1995). Increased aridity and wind conditions enhance aeolian transport (e.g., Bozzano et al., 2002), which can transport detrital carbonate material, coming mostly from North Africa (e.g., Negral et al., 2012; Stumpf et al., 2010) to the [JbM](#). On the other hand, increased moisture and precipitation intensifies rock weathering and riverine discharge, raising fluvial transport of detrital carbonate onto the [JbM](#) (Hodell et al., 2017; Hodell and Channell, 2016). [However](#), the [JbM documents](#) increased detrital carbonate presence is prevalent during [cold and arid](#) rapid millennial-scale [events related with](#) low sea-level stands and IRD events, as documented by Lebreiro et al. (2009), Marino et al. (2011), [Hodell et al. \(2013, 2017\) and Hodell and Channell \(2016\)](#).

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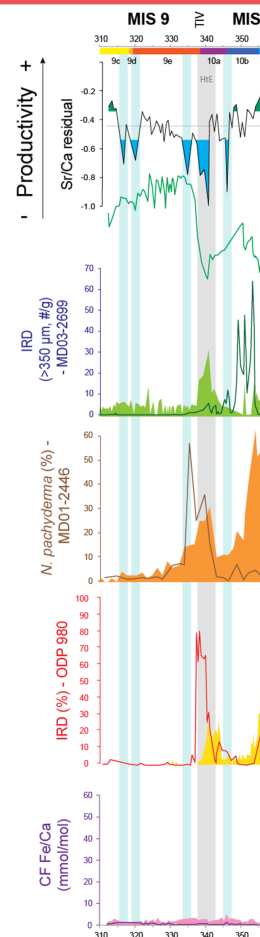
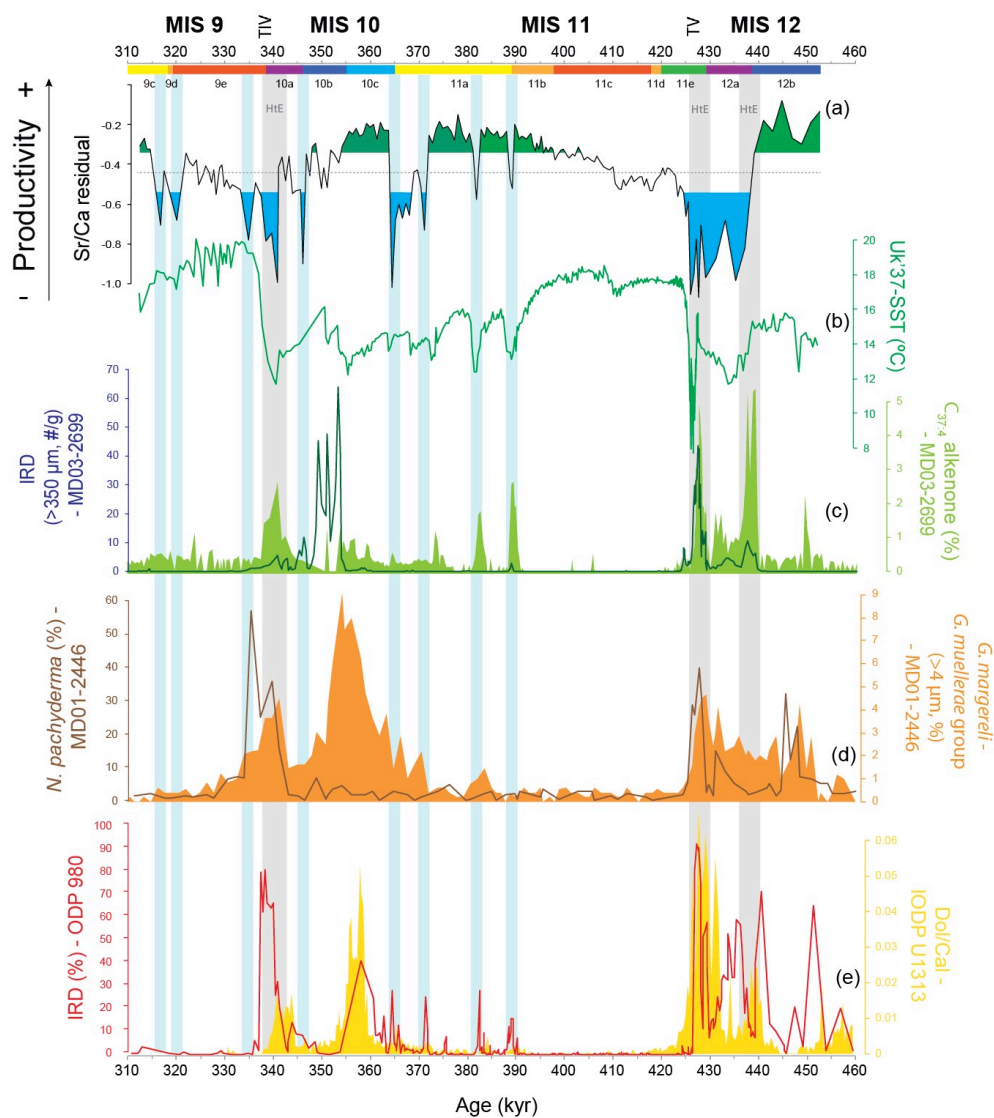
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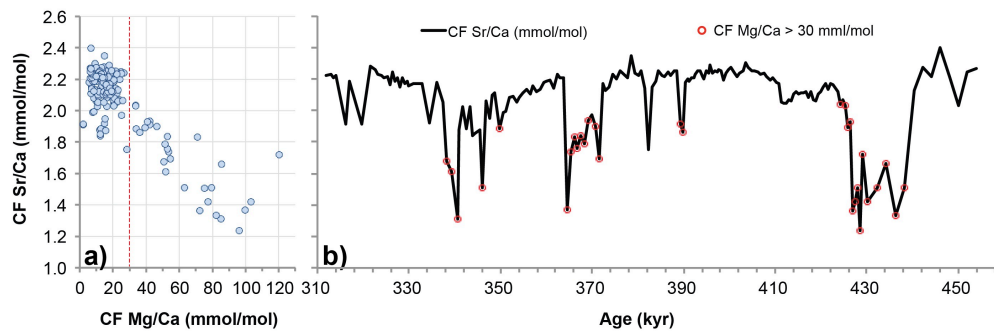
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Figure 4 Coccolithophore paleoproductivity reconstruction with “high” and “low” coccolithophore productivity levels highlighted in green and blue shading, respectively (a) compared to: b) $U^{k'}_{37}$ -based reconstruction of sea surface temperature from core MD03-2699 (Rodrigues et al., 2011); c) ice-rafted debris (IRD) abundance (Voelker et al., 2010) and heptatriatetraene ($C_{37:4}$ alkenone) abundance (Rodrigues et al., 2011), both from core MD03-2699; d) *Neogloboquadrina pachyderma* relative abundance and *Gephyrocapsa marginata* and *G. muelleri* group relative abundance, both from core MD01-2446 (Marino et al., 2014); e) percentage ice-rafted debris (IRD) from ODP Site 980 (McManus et al., 1999) and Dolomite/Calcite ratio from IODP Site U1313 (Stein et al., 2009). Note that the MD03-2699 lithic fragments were counted in a coarser size fraction ($>315 \mu m$) than the standard size fraction ($>150 \mu m$; Hemming, 2004) and thus most likely only record major ice-rafting events at the IbM, as suggested by Marino et al. (2014). Chronology as in Fig. 3. Vertical bars: grey bars correspond to Heinrich-type events (HtE) and blue bars to short-lived events of decreased coccolithophore productivity.

In order to evaluate this potential biasing effect we match the CF Mg/Ca values in a cross-plot with the CF Sr/Ca results. We used CF Mg/Ca because Mg/Ca peaks can be attributed to the presence of increased detrital carbonate, such as dolomite ($CaMg(CO_3)_2$). The Mg/Ca threshold was defined at 30 mmol/mol because from that point onwards both CF Mg/Ca and Sr/Ca show a clear negative relationship, i.e., CF Mg/Ca increases, when CF Sr/Ca clearly decreases (Fig. 5a). Therefore the samples highlighted in Fig. 5b with a red circle were considered possibly contaminated by detrital carbonate. We estimate that (i) 16% of the samples might have been biased by the presence of detrital carbonate and confirm that (ii) such samples are linked to rapid millennial-scale and low sea-level stands, namely during substages MIS 10a, MIS 12a and the respective subsequent terminations V and IV (Fig. 4). Although a possible bias effect exists in such samples, it does not prove that coccolith Sr/Ca ratio and coccolithophore productivity were not indeed significantly lower during those intervals (see Discussion). We have considered the bias from reworked coccoliths contribution negligible because reworked coccolith abundance was generally below 2% not only in our study site (Amore et al., 2012) but also in other IbM sites for the same time interval (Maiorano et al., 2015; Marino et al., 2014).



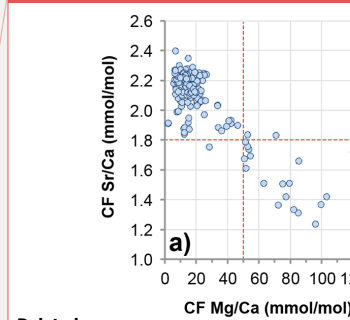
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Figure 5 Detrital carbonate analysis: a) coccolith fraction (CF) Sr/Ca results and CF Mg/Ca cross-plot. Vertical red line delimit threshold at 30 mmol/mol of Mg/Ca. b) CF Sr/Ca samples possibly contaminated with detrital carbonate (red makers) and notably associated millennial abrupt events.

4.2 Coccolithophore productivity results

The CF Sr/Ca ratio results varied between 1.2 and 2.4 mmol/mol (sampling range of 1.16 mmol/mol) with 85 % of the samples falling between 1.8 and 2.3 mmol/mol. The Sr/Ca residual, from now on referred to as coccolithophore productivity (CP), generally mimics the CF Sr/Ca ratio record, and shows minima and maxima at the same levels (Fig. 3). The high CF Sr/Ca ratios found in the record do not coincide with increased abundance of large/Sr-rich coccoliths (Fig. 2) or with higher temperatures (Fig. 3). This supports the statement that our CF Sr/Ca variation reliably records coccolithophore calcification rate and productivity changes. Both MIS 12 and MIS 10 are characterized by higher CP during the first half and lower productivity towards the second half. Large and rapid increases in CP characterize the deglaciations at the end of MIS 12 and MIS 10.

To better visualize and interpret the CF Sr/Ca results, the CP data were divided into three relative intervals. An “intermediate” interval is defined by the mean of the data plus and minus half of the standard deviation. The “high” and “low” CP levels are above and below the intermediate level of CP, respectively (highlighted by green and blue shading in figures, respectively). We would like to stress that our study focuses on the qualitative characteristics of the coccolithophore paleoproductivity record, rather than quantitatively estimating the productivity of coccolithophores.

A preliminary visual inspection of the record already allows the identification of higher amplitude events during glacial substages, MIS 12b and 12a, and MIS 10b and 10a, in comparison to the less variable interglacial substages, MIS 11c and MIS 9e. CP maxima happened during the first half of MIS 12 and MIS 10 and during the transition from interglacial to glacial substages (Fig. 3). Productivity minima, on the other hand, occurred during glacial maximum substages MIS 12a and MIS 10a and at the end of MIS 11a. Marine Isotope Stages 11c and 9e, commonly known as the interglacial substages (Past Interglacials Working Group of PAGES, 2016), mainly show periods of lower amplitude changes within intermediate level of CP when compared to the rest of the record. Rodrigues et al. (2011) mentioned a similar SST pattern, on the same time frame and at the same site, which could imply SST as an important factor affecting CP.

5 Discussion

5.1 Causes for CP change

5.1.1 High CP levels during the transition from interglacial to glacial periods

CP increases steadily from mid MIS 11c until the end of MIS 11b and remained generally high from late MIS 11c to MIS 10c, prior to the glacial substages within MIS 10 and MIS 12. We suggest this increasing CP is a consequence of a gradual

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595 rise in nutrient availability due to either enhanced northerly winds and upwelling strength or due to the decreasing influence
of subtropical waters, substituted by increasing influence of transitional nutrient richer surface waters (i.e. ENACWsp).
During interglacial substages, such as MIS 11c and MIS 9e, the wind stress associated to the upwelling events has been
600 suggested to be lower than during glacial substages in the IbM (e.g., Salgueiro et al., 2010). This is based on the observation
that along with the expansion of the northern hemisphere continental ice sheets, atmospheric and oceanic circulation changed
leading to a narrowed the latitudinal temperature gradient and potentially enhanced northerly winds and upwelling intensity
(Broccoli et al., 2006; Hostetler et al., 1999). Note however, that for upwelling to be intensified during transitions from
interglacial to glacial substages, as well as and during glacial substages, it would have required that the atmospheric
605 circulation pattern governed by the relative position and gradients between high and low pressure centres have remained
similar to today (i.e. interglacial substage). Based on differences in the planktonic oxygen isotope records of cores MD03-
2699 and MD01-2446 (more offshore) (Fig. 1), Voelker et al. (2010) suggested that the higher variability in our core, MD03-
2699, could reflect variations in upwelling of deeper waters into the thermocline. Stronger winds would favour upwelling
610 intensification, which could lead to the upwelling of the deeper, nutrient-rich ENACWsp on the IbM (Fiúza, 1984), as
already suggested for some late Pleistocene periods (Salgueiro et al., 2010, 2014). Still, the transition period is characterized
by a decrease in SSTs most likely already reflecting the southward displacement of the North Atlantic frontal system and,
consequently increasing the influence of ENACWsp origin, more nutrient rich than ENACWst origin, in the surface water of
western IbM. Increased nutrient availability would thus support an increase in productivity of the whole phytoplankton
community (Álvarez-Salgado et al., 1997; Guerreiro et al., 2013), and after depletion of silica by diatoms, coccolithophores
615 species capable of rapid growth, as *Gephyrocapsa caribbeanica* is thought to have been, given its cosmopolitan distribution
and dominance in the sediments (Baumann and Freitag, 2004; Bollmann et al., 1998) could bloom. It is important to refer
that such CP maxima would only be possible if bioavailable Si and Fe were not sufficiently high for diatoms to outcompete
coccolithophores until complete nutrient depletion. Maybe seasonality could have also played an important role here (see
intermediate CP level discussion below). As coccolithophores perform better in stable and continuously nutrient replenished
surface waters, in contrast with diatoms that thrive in dynamical (e.g., upwelling) conditions, (Cermeno et al., 2011), we
620 consider that during the transition to full glacial conditions persistent and increasing nutrient replenishment on the IbM was
more related to the advection of transitional waters of subpolar origin than to increased upwelling. Finally, our high CP
supports the idea that the coccolithophore community was able to better perform under these transitional conditions, despite
colder SST, and possibly more turbulent settings than during interglacial substages. Noteworthy is the fact that this maximal
CP interval was frequently interrupted by abrupt cooling events (Fig. 6) when CP also abruptly decreased. Though detrital
carbonate influence cannot be excluded (see methodology), such CP reductions might indicate that coccolithophore
625 productivity was close to its lowest SST threshold and that abrupt drops in SST immediately resonated in abrupt CP declines.

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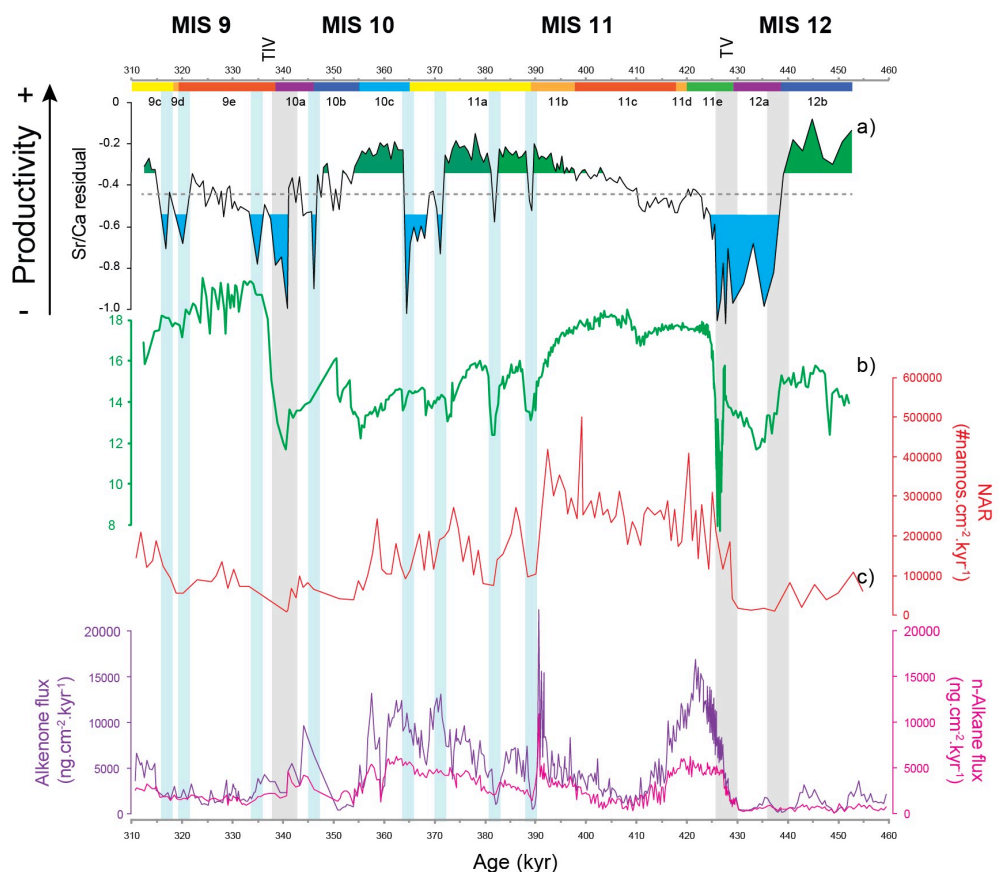


Figure 6 Coccolithophore paleoproductivity reconstruction with “high” and “low” CP levels highlighted in green and blue shading, respectively (a) compared to: b) U^{K}_{37} -based reconstruction of sea surface temperature (Rodrigues et al., 2011), c) nannofossil accumulation rate (NAR) (Amore et al., 2012) and d) total alkenone (AlkAR) and n-alkane accumulation rates (Rodrigues et al., 2011). Chronology as in Fig. 3. Vertical bars: grey bars correspond to Heinrich-type events (HtE) and blue bars to short-lived events of decreased CP.

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5.1.2 Intermediate CP levels associated with deglaciations and interglacial periods.

The rapid CP increase during deglaciation is contemporary with a rapid SST rise (T V and T IV; Fig. 3). The deglacial SST increase and the subsequent particularly stable SSTs during MIS 11c (and most likely MIS 9e) recorded in our MD03-2699 core are linked to the increasing influence and dominance of subtropical oligotrophic waters from the Azores and Iberian Poleward Currents (Rodrigues et al., 2011; Voelker et al., 2010). Several IbM sites support this with increased abundance of warm coccolith taxa (site MD03-2699, Amore et al., 2012 and Palumbo et al., 2013a; IODP Site U1385, Maiorano et al., 2015; site MD01-2446, Marino et al., 2014) and of tropical planktonic foraminifera species *Globorotalia menardii* and *Sphaeroidinella dehiscens* (site MD03-2699, Voelker et al., 2010; site MD01-2443, de Abreu et al., 2005) during the deglaciations and MIS 11c and MIS 9e. Despite the large amplitude change in the CP during the deglaciation, a comparably larger amplitude change is associated with glacial substage MIS 12a and the transitional substage MIS 11b, when SSTs were comparably lower than during MIS 11c (see previous subchapter). Thus, we suggest that during the deglaciation CP increased sharply because of the amelioration of the oceanographic conditions (mostly due to increasing SST) but their productivity was limited to intermediate levels of productivity because of either limited nutrient supply from the prevalent advected subtropical oligotrophic waters or upwelled waters (1), and/or increased competition for nutrients with diatoms (2), which mostly affected the coccolithophore species with opportunistic and fast-growing characteristics (r-selected species) such as *gephyrocapsids*. Such competition with diatoms could be related to upwelling intensity (Cernéjo et al., 2011) during interglacial substages MIS 11c and MIS 9e. During the current interglacial, upwelling only happens in the IbM during the spring-summer months, when winds, mostly driven by trade winds, are strong and predominantly from the north/northwest (Fiúza et al., 1982; Fiúza, 1983; Mason et al., 2006). Given the current variability in the western IbM (Alvarez et al., 2008; Aristegui et al., 2009) and its dependency on changes in the regional atmospheric circulation, especially related to the latitudinal migration of the subtropical front and to the dynamics of the Azores anticyclonic cell (Aristegui et al., 2009; Fiúza et al., 1982; Mason et al., 2006), it is difficult to assert what has been the wind regime and upwelling seasonality during the deglaciations. However, when assessing the hypothesized upwelling strength between interglacial and glacial substages, and in the event of lower nutrient availability for the rest of the year, coccolithophores would be easily outcompeted by diatoms, as in the upwelling event of the current interglacial (e.g. Abrantes and Moita, 1999). These evidences suggest that irrespective of the upwelling regime that influenced seasonal overall nutrient availability and phytoplanktonic productivity, the intermediate levels of CP could be explained by a competitive advantage of diatoms upon sufficient delivery of Si and Fe of terrigenous input, either by increased aeolian input or riverine discharge. Since changes in the SST and nutrient availability are not only promoted by fluctuations in the upwelling regime (1) and changes in the advection of the dominant water masses (2) in the IbM, we also considered changing seasonality, i.e. changes precipitation (3) and arid plus windy (4) conditions to have played a role in determining CP. It is well known that the addition of Fe into the surface ocean positively affects the overall phytoplankton community (Blain et al., 2004; Martin et al., 1990). And diatoms require more Si and Fe (e.g., Merico et al., 2004), whereas coccolithophore blooms are thought to be linked with Si

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and Fe depletion (Merico et al., 2004; Tyrrell and Merico, 2004), giving coccolithophores a competitive advantage in a Si and Fe depletion scenario with plenty other nutrients, such as nitrate and phosphate. Thus, under fluctuating nutrient availability and increased bioavailability of Si and Fe (Capellacci et al., 2013), diatoms could have outcompeted coccolithophores via nutrient competition, limiting coccolithophores growth to intermediate levels of productivity, after diatoms bloom. Periods of strengthened wind regime and aridity are generally limited to glacial substages and stadials (Desprat et al., 2009, 2017; Hodell et al., 2013b; Oliveira et al., 2016; Sánchez Goñi et al., 2016). But, Rodrigues et al. (2011) interpreted the synchrony between high terrigenous input (concentration of n-alkanes) and alkenone concentration in core MD03-2699 (Fig. 6d) with intensified dust export from North Africa (based on Fe counts at Site 958 off western North Africa, Helmke et al., 2008), and as indicative of a strong wind regime in the IbM during the deglaciation and early phase of MIS 11c. However such high Fe counts evidencing strong winds are referred by Helmke et al. (2008) for the deglaciation, but not for MIS 11d and early MIS 11c, where the same study suggests a humid period in North Africa associated with increased precipitation and a widespread vegetation cover. So during the deglaciation such strong winds might have been sufficient to replenish the surface waters with Si and Fe, allowing diatoms to bloom (Abrantes, 2000; Capellacci et al., 2013) and outcompeting coccolithophores. And, in early MIS11c Si and Fe could have also come into the IbM through riverine discharge. Under full interglacial conditions, pollen records evidence the development of temperate and humid deciduous forest in northwestern Iberia (Desprat et al., 2007), which during MIS 11c coincided with the North African humid period described by Helmke et al. (2008). Based on the increased abundance of Mediterranean forest taxa observed in southwest IbM IODP Site U1385, Oliveira et al. (2017) associated climate during MIS 11c with the present-day SW Iberian climate, i.e. with pronounced seasonality between warm/dry summers and cool/wet winters. The warmer/drier summers, tied to the northeastwardly expansion of the Azores subtropical High, associated with the descending branch of the Hadley cell (Lionello et al., 2006; Oliveira et al., 2017), could have enabled dust flux to the western IbM. During winters, on the other hand, nutrients in the surface waters could have been replenished by increased precipitation and associated river run-off, in agreement with the expansion of temperate humid forest. So, during interglacials, our theory that periods when CP seemed limited to intermediate levels, are linked to conditions of increased terrigenous input, either aeolian or riverine, that allowed diatoms to outcompete coccolithophores is also valid.

5.1.3 Low CP associated with abrupt cooling events and glacial MIS 12a

CP shows consistent minima along the record that, with the exception of glacial maximum stage MIS 12a and the ~10 kyr interval at the transition from MIS 11a to MIS 10c, are of short duration and related to abrupt climate change events. Thus, most of the minima coincide with the presence of colder and less saline surface waters that resulted from meltwater incursions onto the IbM evidenced either by the increased IRD content, or higher percentages of C_{37:4} alkenones, concomitant with higher abundances of coccoliths belonging to *Gephyrocapsa margaritae*-*G. muelleriae* (>4 µm), a cold/low salinity water proxy in the IbM (Fig. 4; Marino et al., 2014; Rodrigues et al., 2011). The most extreme of these events are the HtE (Stein et al., 2009; Fig. 4), with the ones during TIV and TV corresponding to the terminal stadial events of Hodell et al. (2015). The

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995 associated cooling brought the SSTs nearly to local glacial levels (Fig. 4; Rodrigues et al., 2011). Similar to their counterparts during the last glacial cycle, the meltwater events resulted in an accentuated decrease or nearly elimination of the Atlantic Meridional Overturning Circulation and in a southwards displacement of the North Atlantic frontal system with the subpolar/arctic front moving into IbM latitudes (Alonso-Garcia et al., 2011; Hodell et al., 2008; McManus et al., 1999; Rodrigues et al., 2011, 2017). This led to the advection of polar and subpolar water masses onto the IbM evidenced by the increased percentage of polar planktonic foraminifera *Neogloboquadrina pachyderma* in the nearby core MD01-2446 (Fig. 4; Marino et al., 2014) and in cores off Galicia (Desprat et al., 2007, 2009). The abrupt CP drops at 438 ka and at 340 ka prior to and during TV and TIV, respectively, are both related to HtE (Fig. 4). As previously mentioned, during HtE detrital carbonate might have negatively biased our CP. However, detrital carbonate might have acted as a low productivity signal amplifier instead of attributing false low CP in such intervals. An extreme decline in CP during the TV HtE/terminal stadial event was also observed at IODP Site U1313 in the central North Atlantic basin (Cavaleiro et al., 2018). During those events the abrupt SST decrease coupled with increased turbidity and blockage of sunlight (due to melting icebergs and/or sea ice cover) would have decreased the coccolithophores' ability to survive. These conditions resemble those in the present-day polar domain of the Norwegian-Greenland Sea (Baumann et al., 2000). Here, a less diverse coccolithophore community was observed with a slightly diminished coccolithophore population, which could well support the abrupt decreases detected in our CP record. Additional CP minima are detected at 389 ka, 382 ka, 371 ka, 364 ka, the latter dropping to HtE levels (Fig. 4), 346 ka, 334 ka, 319 ka, and 316 ka. Despite no significant concomitant IRD peaks in core MD03-2699, these decreases are associated with increases in the abundance of the C_{37:4} alkenone and colder SST (Fig. 4; Rodrigues et al., 2011) and the presence of *N. pachyderma* and *G. margereli-G. muelleri* (>4 µm) in core MD01-2446 (Fig. 4; Marino et al., 2014), testifying a rapid change to significantly colder and less saline conditions at this latitude. The four older drops in CP happened concomitantly with abrupt increases in *Neogloboquadrina pachyderma* abundance at ODP Site 980 (55°29'N 14°42'W) in the eastern North Atlantic (Fig. 4; McManus et al., 1999; Oppo et al., 1998), placing them in a broader spatial scale of climate deterioration. Thus, we conclude that the IbM was subjected to drastic surface water changes that led to a coccolithophore community decrease in the IbM and generated abrupt decreases in our CP record. Finally, the reduced productivity during MIS 12a at the location of core MD03-2699 is in stark contrast to the open ocean record of IODP Site U1313 (Cavaleiro et al., 2018) and other past glacial periods in the IbM such as MIS 2 and MIS 6 (Salgueiro et al., 2010, 2014), all of which point to increased productivity during the glacial maxima. Furthermore, such prolonged low CP is also not observed during glacial maximum MIS 10a, when productivity at site MD03-2699 remained at intermediate levels between abrupt cold events (Fig. 4). However, the prolonged presence of (sub)polar waters in the eastern North Atlantic throughout MIS 12a is backed up by the IRD record and persistent high percentages of *N. pachyderma* (Oppo et al., 1998; McManus et al., 1999) at ODP Site 980 and in core MD01-2448 in the Bay of Biscay (Toucanne et al., 2009). It is thus likely that such low temperature, and eventually salinity, water masses also affected the IbM, hampering upwelling

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1065 (enhanced upwelling is seen as cause for increased productivity at the IbM during glacial maxima; Abrantes et al., 2000; Salgueiro et al., 2010, 2014) and potentially leading to a low CP.

5.2 Evidence from different coccolithophore productivity proxies and conceptual models

1070 Previous studies in the IbM used coccolith-derived proxies, such as coccolith assemblages, nannofossil accumulation rates (NAR), and total alkenone fluxes (produced mostly by coccolithophores) to reconstruct changes in coccolithophore paleoproductivity (Amore et al., 2012; Maiorano et al., 2015; Marino et al., 2014; Palumbo et al., 2013). These traditional proxies depend not only on the supply of coccoliths or organic compounds but also on dilution by minerals and other sediment constituents and on changing preservation conditions (Rullkötter, 2006). As the CF Sr/Ca ratio is an independent proxy, i.e. independent of sedimentation and accumulation rates, it offers a new perspective into CP dynamics and the opportunity to compare coccolithophore growth/calcification rate (referred here as coccolithophore productivity) with other components of the system. In a simple time-series comparison of our CP data with the NAR or the AlkAR records we can already confirm that their relationship is not straightforward (Fig. 6). While in some intervals CP, NAR and AlkAR show similar trends, in others they diverge.

1080 As mentioned previously, several studies have hypothesized a relevant role of coccolithophores on the global carbon and carbonate cycles and on the glacial-interglacial dynamics (Duchamp-Alphonse et al., 2018; McClelland et al., 2016; Omta et al., 2013; Rickaby et al., 2007; Saavedra-Pellitero et al., 2017). Yet, the inclusion of biogeochemical processes related to coccolithophores in Earth System models is lacking although they play a role in both the organic (soft tissue) and the carbonate counter pump (through calcification). Coccolith carbonate also has a ballasting effect, enhancing the C_{org} export efficiency (Armstrong et al., 2001; Francois et al., 2002) but the relative proportion of exported particulate organic carbon to inorganic carbon are difficult to predict and model (Barker et al., 2006; Ridgwell and Zeebe, 2005). Therefore, the respective and relative importance of these mechanisms is still poorly understood and the past and future net impact of calcification by coccolithophores on $atmCO_2$ is still uncertain (Lawton et al., 2003). Among the difficulties in modelling coccolithophores' responses are the complex interactions between coccolithophore productivity and growth rates and nutrient dynamics, non-calcifiers competition, seasonality and their response to upwelling/non-upwelling conditions.

1090 We suggest that the contemporaneous coupling or decoupling of the different coccolithophore productivity proxies reflect changes in the carbon and carbonate system and conceived conceptual models of the carbon and carbonate cycles in the IbM (Fig. 7).

1095 During the MIS 12 **deglaciation** all coccolithophore proxies show similar trends. This deglaciation is marked by clear rapid increases in CP, NAR and AlkAR accompanied by a rapid, high amplitude rise in SST, characteristic of terminations (e.g. Rodrigues et al., 2017). The increasing CP is linked to the growing influence and dominance of subtropical oligotrophic waters from the Azores and Iberian Poleward Currents (SST evidence) combined with a moderate to strong wind regime and consequent upwelling (Voelker et al., 2010). Based on paleoclimatic evidence deglacial hinterland conditions in western Iberia and NW Africa were arid and resulted in increased (Saharan) dust flux (Desprat et al., 2007; Helmke et al., 2008;

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Hodell et al., 2013a, 2015; Rodrigues et al., 2011), which would supply high amounts of Fe and Si to the IbM. We suggest that this increased nutrient combination fertilized the non-calcifying phytoplankton component, namely diatoms (Duchamp-Alphonse et al., 2018; Helmke et al., 2008; Meckler et al., 2013; Rodrigues et al., 2011; Thomson et al., 2000), attributing them a competitive advantage over coccolithophores, outcompeting them (e.g. Abrantes and Moita, 1999; Balch, 2004; Guerreiro et al., 2013). The combined increased surface productivity with both diatoms and coccolithophores blooming would likely allow for an increase in the soft-tissue pump into the deep-ocean. During the **deglaciation and early interglacial phases**, the augmented, though only to intermediate levels, coccolithophore productivity was associated with higher calcification rates, and thus CP, that resulted in a higher NAR. The higher CaCO₃ production at the surface might have led to an increased CaCO₃ and C_{org} export efficiency through the ballast effect of CaCO₃ (e.g. Armstrong et al., 2001; Francois et al., 2002). This could explain the enhanced AlkAR as well as NAR at our study site for the deglaciations and early MIS 11c and MIS 9e, though with significantly different amplitudes.

During **mid interglacial MIS 11c and MIS 11b** (410 kyr to 390 kyr) and MIS 9e, all coccolithophore productivity proxies steadily increase, most likely associated with decreasing competition with diatoms for nutrients (see previous discussion section). However the quite different AlkAR records for the **deglaciation and early MIS 11c** (428 kyr to 410 kyr) in comparison with **the rest of MIS 11c and MIS 11b** (410 kyr to 390 kyr) suggest a change in the phytoplanktonic community, favouring coccolithophores in detriment of diatoms. We suggest that while diatoms were receiving a surplus replenishment of Si and Fe (as discussed in section 5.1.2) the soft tissue pump became more efficient because more organic particulate matter and particulate inorganic matter was present to form fast sinking aggregates (Armstrong et al., 2001; Francois et al., 2002) allowing also for increasing amount of alkenones to be exported and accumulated. We hypothesize that the timing for the rapid decrease in AlkAR could actually coincide with the decrease in the surface particulate organic matter as a consequence of the decreasing diatoms productivity and surplus replenishment of Si and Fe, which ceased by 410 kyr, when AlkAR reach their minimum. This could have important implication in past CO₂ fluxes from the atmosphere into the deep-ocean, if indeed the soft-tissue pump increased its efficiency due to this combination of factors.

From **mid MIS 11c to the end of MIS 11b** (410 kyr and until 390 kyr), all coccolithophore productivity proxies increase and we suggest that this coupled signal reflects the coccolithophore productivity had a diminished competition with diatoms for nutrients, and an associated less efficient soft-tissue pump in comparison to the **deglaciation and early MIS 11c**.

Based on the evidence from IODP Site U1313 in the open mid-latitudinal North Atlantic (Cavaleiro et al., 2018), increases in coccolithophore calcification rate and productivity do not necessarily have to be a consequence of enhanced upwelling but can just as likely be related to the southward migration of the high productivity/high coccolithophorid belt (Cavaleiro et al., 2018; McIntyre et al., 1972; Villanueva et al., 2001). This band, currently located between 45 °N and 55 °N (Antoine et al., 1996; Henson et al., 2009; Longhurst et al., 1995), is associated with the convergence zone between the subpolar and subtropical gyres (McIntyre et al., 1972). Along with the southward expansion of the subpolar gyre during periods of slowdown of the North Atlantic's thermohaline circulation (Alonso-Garcia et al., 2011; Rodrigues et al., 2017; Wright and Flower, 2002), the high productivity band moved into the latitudes of the western IbM. So, the (gradual) displacement of this

high coccolithophorid belt could explain the rapid rise in CP productivity during MIS 11b, and associated maxima during MIS 11a, MIS 10c and MIS 9c when all three records, CP, NAR and AlkAR, show contemporary increases (Fig. 6). Another factor affecting the signal preservation during the glacial periods and abrupt cold events of MIS 11b to MIS 11a and of MIS 9d to MIS 9c could be shifts in the conditions prevailing at the sea-floor. Whereas site MD03-2699 was bathed by NADW when the North Atlantic's thermohaline circulation was strong, i.e. during the interglacial substages and the interstadials of MIS 11a, the North Atlantic Deep Water was replaced by Mediterranean Outflow Water (MOW) during times of a weak thermohaline circulation (stadials of MIS 11b and MIS 11a; glacial MIS 12a and most of MIS 10) (Voelker and Lebreiro, 2010). Periods of MOW presence were associated with increased bottom current activity at site MD03-2699, especially during MIS 12a, when winnowing of finer particles could have contributed to the low NAR and AlkAR (Fig. 6). On the other hand, the less oxygenated MOW would induce less C_{org} remineralization what could have played a role in the AlkAR maxima occurring during late MIS 11a

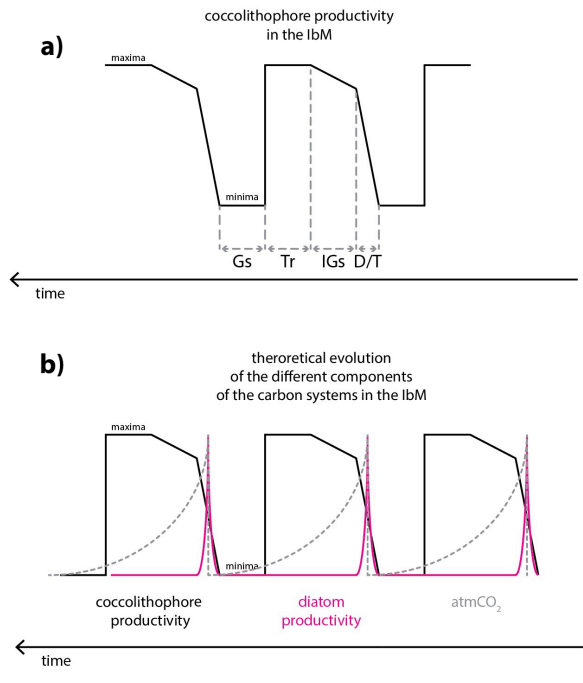
5.3 Further research suggestions

The CP reconstruction for the mid-Brunhes interval at site MD03-2699 yielded important findings, but also raised issues that should be explored in the future. Extending this research into other interglacial substages, could help disentangle the phytoplankton community response to upwelling intensity, namely if both diatoms and coccolithophores increase or if one decreases in detriment of the other with changing upwelling intensities. This would offer further insights into the role of particulate organic vs. inorganic production in the surface ocean in an upwelling area such as the IbM. Further attention should also be given to the role of seasonality in CP and diatom productivity dynamics the IbM, which could potentially be disentangled by using a direct land-sea correlation, namely evidence from pollen records. Given the discrepancy between our and other surface productivity and export estimations for glacial periods on the IbM (Abrantes, 1992; Salgueiro et al., 2010, 2014) it would be interesting to assess coccolithophore productivity in late or early Pleistocene glacial stages to assess if our observation of low CP are restricted to mid-Brunhes conditions.

Additional research, specially incorporating the gained information into carbon and carbonate cycle models, could help to better predict the role of the IbM and other (eastern boundary) upwelling systems on glacial-interglacial $atmCO_2$ changes. Did the coupled high productivity of non-calcifiers and calcifiers during deglaciations operate as sink of carbon or did the carbonate counter pump act to balance or even counteract the amplified soft-tissue pump?

Finally, Omta et al. (2013) presented a potential role of coccolithophores in the glacial-interglacial cycles of $atmCO_2$ and based their experiments on prey-predator models with the characteristic spiking predator (like coccolithophores) and a saw tooth shape curve of the prey behaviour (like alkalinity, which is positively and directly correlated with $[CO_3^{2-}]$). Despite the undeniable direct link between calcifying phytoplankton and ocean alkalinity (e.g. Ridgwell and Zeebe, 2005), their model with the spikes of enhanced productivity at deglaciations/terminations is actually resembling more the non-calcifiers behaviour in the IbM and off Africa, namely diatoms (Meckler et al., 2013; Thomson et al., 2000), than of coccolithophores, as our record shows. Coccolithophores productivity in the IbM margin seems to rise abruptly during the deglaciations,

1195 followed by a constant level or slight increase during the interglacial substages, and then to steadily increase further during the progressive global cooling towards the glacial maximum only to then abruptly decrease under full glacial conditions (Fig. 7).



1200 **Figure 7** Theoretical curves of the carbon cycle components in the IbM: (a) hypothesized coccolithophore paleoproductivity for the IbM for the studied interval; (b) Theoretical diatom productivity behaviour, with the characteristic spike at terminations (Meckler et al., 2013), and atmCO₂ changes, with the characteristic sawtooth shape. Note that the intervals were drawn with no specific duration. Gs, Tr, IGs and D/T stand for Glacial substage, Transition from interglacial to glacial substages, Interglacial substages and deglaciation/transition, respectively.

1205 We hope that this information could be included in future modelling experiments, with both coccolithophore and diatom components of the dynamic phytoplanktonic community, to better describe the role of each functional group in the carbon and carbonate cycles in this upwelling system, and with possible implications for other eastern boundary upwelling systems. We urge climate scientists to research the combined responses of the different components of the phytoplanktonic community, namely calcifiers and non-calcifiers. Much research has been conducted exploring only the productivity changes

of one of these functional phytoplanktonic groups. Since the global ocean phytoplanktonic community with potentially greater impact on atmCO₂ fluxes is mostly composed of diatoms and coccolithophores in either the open ocean or upwelling areas, it is important to understand how the phytoplankton community dynamics changed in the past.

6 Conclusions

In this study we applied the CF Sr/Ca ratio to reconstruct CP during the mid-Brunhes interval of MIS 9 to MIS 12 and discussed the different factors controlling CP in the western IbM. We found that the long-term coccolithophore productivity was primarily controlled by changes in the paleoceanographic surface conditions, namely the origin of the predominant surface waters and their nutrient content. CP maxima were found during the transition from interglacial to glacial substages and associated with increasing presence of nutrient-rich transitional surface waters. The CP maxima during this transition were, however, frequently interrupted by abrupt decreases in CP linked to abrupt cooling events (HtE) and growing predominance of polar and subpolar surface waters, such as during glacial maximum substages. The deglaciations and during early MIS 11c were marked by intermediate CP linked to increased influence of subtropical, nutrient-poorer waters during winter, combined with increased terrigenous input of nutrients and moderate to strong upwelling during summer; conditions that are hypothesized to have attributed to a competitive advantage of diatoms over coccolithophores.

Using a multiproxy approach, gathering CP (surface production signal) and AlkAR and NAR (bottom export and accumulation signals) we infer conceptual models of the carbon and carbonates cycles in the IbM during different periods and suggest future research questions to be tested through modelling. We hope that our new data can instigate Earth system modellers to look deeper into constraining the role of coccolithophores in the regional and global carbon and carbonate cycles, especially in the eastern boundary upwelling systems of the world.

Data availability. The data used will be available from the Pangaea data repository <https://www.pangaea.de>

Author contribution. CC and AV designed the study. CC prepared the samples and analysed them under the supervision of HS. CC drafted the paper and all authors contributed for the discussion and to the final version.

Competing interests. The authors declare that they have no conflict of interest.

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the University of Oviedo. CC would also like to thank Dulce Oliveira, Fátima Abrantes, Filipa Naughton and Teresa Rodrigues for the helpful comments to the manuscript. [We thank Tom Dunley Jones and an anonymous reviewer for their constructive reviews that help greatly to improve the manuscript.](#)

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This signal can, however, be removed from the Sr/Ca record using an independent sea-surface temperature (SST) reconstruction, so that the component of variation due to growth rate remains as residual (e.g. Cavaleiro et al., 2018; Mejía et al., 2014; (e.g. Cavaleiro et al., 2018; Mejía et al., 2014; Saavedra-Pellitero et al., 2017). Also, despite the variations among species in the amount of Sr introduced into its calcite (with larger and more heavily calcified coccoliths generally having a higher Sr content than smaller and lighter coccoliths (Fink et al., 2010; Stoll et al., 2007)), it has been demonstrated that CF Sr/Ca ratios are not primarily controlled by variations in coccolith assemblage in the modern ocean (Barker et al., 2006; Stoll and Schrag, 2000). It is thus reasonable to conclude that most CF Sr/Ca ratio records reflect long-term changes in coccolithophore production and calcification. With temperature and assemblage effects considered, the SST corrected CF Sr/Ca curve (or residual curve) can be expected to mostly

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This study reconstructs coccolithophore productivity from the CF Sr/Ca ratio record at Site MD03-2699, retrieved from the western Iberian margin (IbM) (Fig. 1), in an eastern boundary upwelling system. We aim to characterise long-term changes in coccolithophore productivity in such a system, where their behaviour in the past remains unknown. We explore different climatic scenarios during the Mid

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, focusing on the interval from Marine Isotope Stage (MIS) 12 to MIS 9, and evaluate the main factors influencing coccolithophore productivity. This interval was a critical time of important global climate change when, after the mid-Pleistocene transition, glacial-interglacial cyclicity became more stable at the periodicity of ca. 100 kyr (e.g. Berger and Wefer, 2003). Our study period includes harsh glacial periods such as MIS 12 and the prolonged MIS 11c interglacial period. The MIS 11c interglacial is

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, at both orbital and sub-orbital time scales. We further compare our proxy with classical coccolithophore productivity proxies, such as nannofossil accumulation rate (NAR) and total alkenone fluxes.

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As previously mentioned, to correctly interpret the CF Sr/Ca ratio proxy it is important to account for changes in coccolith assemblage and SST through time.

Our study is placed within the *Gephyrocapsa caribbeanica* acme (Baumann and Freitag, 2004; Bollmann et al., 1998) between MIS 14/13 and MIS 8 when this species dominated ($\geq 50\%$) the coccolithophore community globally. This species' governance minimizes the bias of the CF Sr/Ca data due to significant coccolith assemblage changes. Moreover, coccoliths belonging to the genus *Gephyrocapsa* dominated the assemblage in core MD03-2699 during the entire studied period with an average of 97% (Fig. 2; Amore et al., 2012). The sum of the remaining coccoliths, containing the larger and Sr-rich coccoliths *Calcidiscus leptoporus*, *Coccolithus pelagicus* and *Helicosphaera carteri* averaged 2.8 % (maximum 8.7%, standard deviation 1.6%). Their contribution to the total Sr of the coccolith fraction is therefore negligible when compared to the *Gephyrocapsids*' contribution.

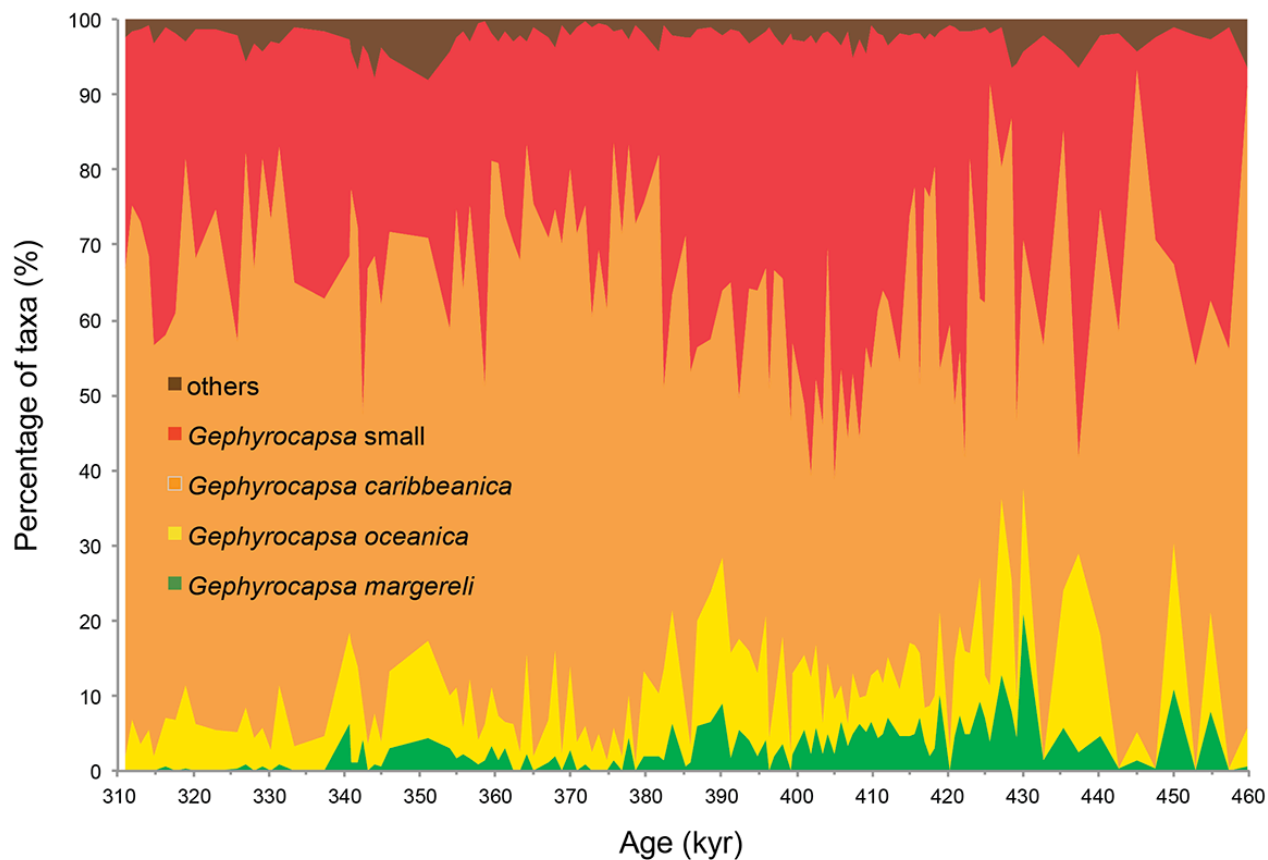


Figure 2 Composition of coccolith assemblages in core MD03-2699 based on Amore et al. (2012). Note that only a minor percentage of coccoliths belong to groups other than the dominant *Gephyrocapsids*.

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3.4 Time-series analysis

Using the outcome of the harmonic analysis of the CF Sr/Ca residual record in the SPECTRUM program (Schulz and Stattegger, 1997), we extracted frequencies lower than 37 kyr from the coccolithophore productivity record (i.e. residuals of eccentricity and obliquity whose occurrence in the time series is not frequent enough for reliable results). The filtered record was then used for the spectral and cross-spectral analysis in the programs REDFIT and SPECTRUM, respectively (Schulz and Mudelsee, 2002, Schulz and Stattegger, 1997), to establish statistically significant leads/lags between the coccolithophore productivity record and seasonal insolation.

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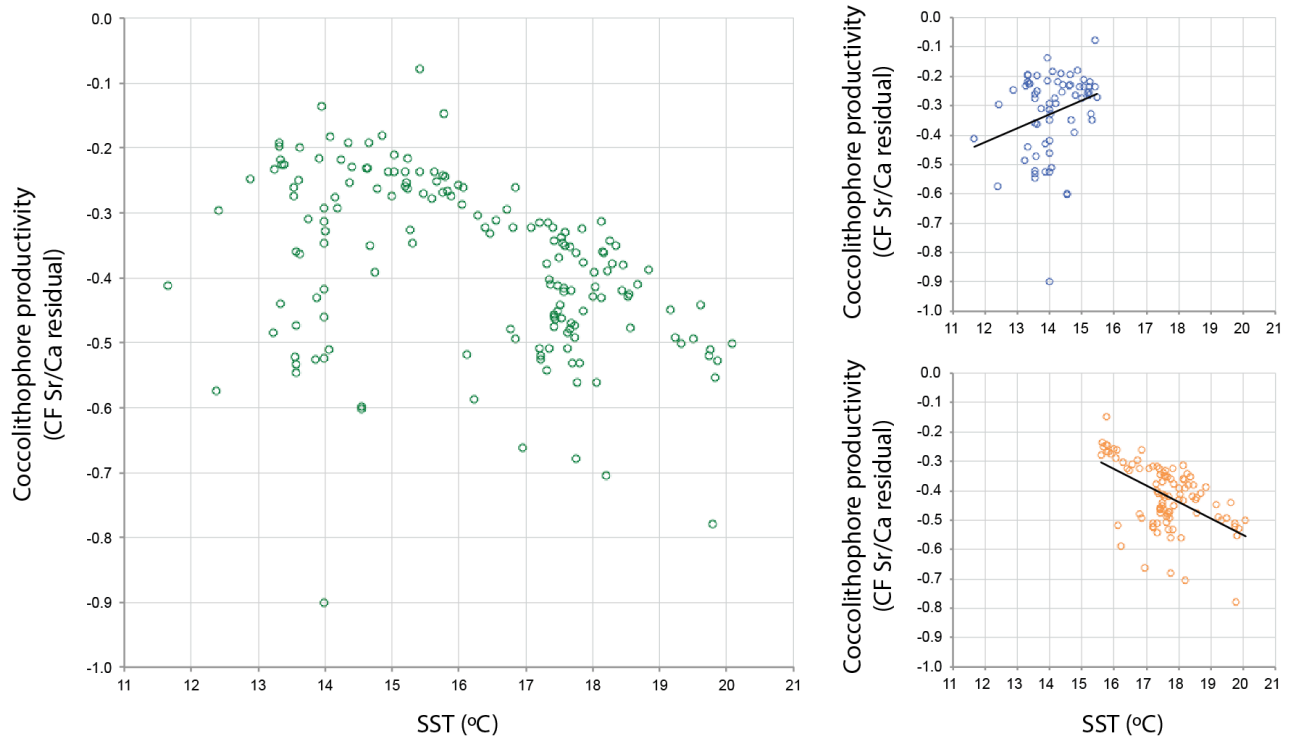
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Coccolithophore productivity

We therefore firstly evaluated the relationship between coccolithophore productivity and SST changes. The relationship between SST and coccolithophore productivity is characterised by a weak negative correlation ($r=-0.4$, $p<0.01$). However, the cross-plot seems to show an inflexion around 15.5 °C (Fig. 7) and, by separating the different sets of samples under the different temperature ranges (below and above 15.5 °C), we obtained very different correlation results. Increasing temperatures until 15.5 °C seem to have a positive effect on coccolithophore productivity (though with a low positive correlation; $r=0.27$, $p<0.05$), whereas increasing temperatures above 15.5 °C seem to have a negative effect on coccolithophore productivity (with a medium negative correlation; $r=-0.54$, $p<0.01$). Though it is accepted that increased temperature generally enhances coccolithophores growth (e.g. Sett et al., 2014), our results demonstrate that the relationship between coccolithophores productivity and SST is not straightforward and should be explored further in the future and that other factors, besides temperature, must have affected coccolithophore productivity in the western IbM. In an upwelling region like the IbM a potential factor could be nutrient concentrations because the upwelled waters in general have temperatures between 13 and 16 °C (e.g., Fiúza et al., 1998).



Correlation Coefficients - Cocolithophore productivity (CF Sr/Ca residual) vs SST (°C)

	<i>Sr/Ca residual vs. SST (°C)</i>	<i>Sr/Ca residual vs. SST (°C)</i>	<i>Sr/Ca residual vs. SST (°C)</i>
R	-0.4	0.27	-0.54
<i>R Standard Error</i>	5.17E-3	0.01	7.3E-3
<i>t</i>	-5.52	2.22	-6.32
<i>p-value</i>	1.29E-7	0.03	7.8E-9
<i>H0 (5%)</i>	<i>rejected</i>	<i>rejected</i>	<i>rejected</i>
<i>No# of valid cases</i>	165 (all valid samples)	66 (samples SST < 16 °C)	99 (samples > 16 °C)

Figure 7 Cocolithophore paleoproductivity and SST Pearson correlation results: all valid samples (green), samples below 15.5 °C (blue) and samples above 15.5 °C (orange). Person correlation computed with StatPlus:mac, AnalystSoft Inc. - statistical analysis program for macOS®. Version v7. See <http://www.analystsoft.com/en/>

5.1 Causes for cocolithophore productivity change

5.1.1

terminations and interglacial periods

The abrupt SST rise during the

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Accordingly, during MIS 9e and MIS 11c, the abundance of warm coccolith taxa increased (Amore et al., 2012; Maiorano et al., 2015; Marino et al., 2014; Palumbo et al., 2013a) as well as the abundance of tropical planktonic foraminifera species in cores MD01-2443 (de Abreu et al., 2005) and MD03-2699 (presence of *Globorotalia menardii* and *Sphaeroidinella dehiscens*; Voelker et al., 2010). These evidences point to the prevailing presence of subtropical oligotrophic waters during the later phase of the deglaciation and across interglacial MIS 11c. As a consequence of the limited nutrient supply, coccolithophore productivity, especially the opportunistic and fast-growing species, could have been limited to intermediate levels during MIS 11c.

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Besides changes in the nutrient availability

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coccolithophore productivity.

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Also, based on evidence from the late Pleistocene (e.g. Margari et al., 2014), the transition from MIS 12 to MIS 11 is likely also marked by a transition from very cold winters and windy conditions in the western and southern Iberian margin to warmer conditions with increased winter precipitation (Desprat et al., 2009; Tzedakis et al., 2009). Higher humidity and precipitation would increase continental weathering and transport of silica and iron to coastal areas. And indeed the coccolith fraction, in which the Sr/Ca ratio was measured, also shows increased silica and iron contents when compared to calcium (CF Si/Ca and CF Fe/Ca; Fig. 4), especially during deglaciation and the beginning of MIS 11c. Also, CF Si/Ca and CF Fe/Ca are intricately connected ($r=0.95$, $p<0.01$), and are both negatively correlated with coccolithophore productivity ($r=-0.44$, $p<0.01$ and $r=-0.49$, $p<0.01$, respectively). This could indicate that even if iron, likely brought by dust originated in the Sahara desert, might have played an important fertilizing effect for the overall phytoplankton community offshore the IbM (Blain et al., 2004) in the past, diatoms most likely outcompeted coccolithophores, limiting their productivity due to increased competition for nutrients, when silica was in sufficient amount not to limit diatom blooms. All of these evidences suggest that irrespective of a moderate to strong upwelling regime, which increased the overall nutrients availability and phytoplanktonic productivity, only sufficiently higher the amounts of silica and iron introduced by increased wind and aridity or by increased precipitation, would lead to diatom blooms and coccolithophores to be outcompeted, most likely explaining the intermediate level of coccolithophore productivity.

5.1.2 High coccolithophore productivity levels associated with the transition from interglacial to glacial periods

Coccolithophore productivity increases steadily from mid MIS 11c until the end of MIS 11b and remained generally high from late MIS 11c to MIS 10c. Although the increasing trend in coccolithophore productivity coincides with a decreasing trend in SST from 403 to 390 kyr (which returned a high negative correlation, $r=-0.88$, $p<0.01$), the high productivity interval, from 398 kyr to 354 kyr, seems disconnected from SST influence ($r=0.16$, $p=0.2$).

The cooling of the SST during the transition from interglacial substages to glacial substages is related to the build-up of ice sheets on the continents and the subsequent changes in atmospheric and oceanic circulation, which subsequently resulted in a strength decline of the AMOC (e.g., McManus et al., 1999; Voelker et al., 2010), also evidenced by the decreasing trend in bottom water ventilation (e.g., Martrat et al., 2007). During interglacial substages, such as MIS 11c and MIS 9e, the wind stress associated to the upwelling events is hypothesized to be lower than during glacial substages (e.g.

Whereas, during the transitions from interglacial to glacial substages, the narrower latitudinal temperature gradient caused by the expansion of the northern continental ice sheets and the associated location shifts in the North Atlantic frontal system, is hypothesized to have increased the northerly winds and lead to more intense upwelling or wind related mixing of the upper water column. As a consequence, higher turbulence would replenish the ocean surface with nutrients.

Because of the differences in the planktonic oxygen isotope records between cores MD03-2699 and MD01-2446 (more offshore) Voelker et al. (2010) suggested that the high variability in the closer to shore core MD03-2699 could reflect variations in upwelling of deeper waters into the thermocline. Indeed, Fiúza (1984) had already proposed that variations in wind stress could lead to the upwelling of different water masses.

Enhanced northerly wind stress could have intensified the upwelling, both in strength (upwelling deeper and nutrient-rich waters) and in distance to shore (reaching further offshore than today). Increased nutrient availability would thus support the whole phytoplankton community and decrease diatoms' and coccolithophores' competition for nutrients, especially for those coccolithophores species capable of rapid growth in a nutrient-rich environment, such as the *Gephyrocapsa caribbeanica* is thought to have been, given its cosmopolitan distribution and dominance in the sediments (Baumann and Freitag, 2004; Bollmann et al., 1998). Note that this would only be possible given that bioavailable silica and iron were not sufficiently high to allow diatoms to outcompete coccolithophores, as seen before, and as the CF Si/Ca seem to show, with quite low and constant levels during the coccolithophore productivity maxima, only interrupted by a period of increased CF Si/Ca and abrupt decrease in coccolithophore productivity, from ~375 to 365 ka (Fig. 4). The coccolithophore productivity maxima during the transition from interglacial MIS 11c to glacial MIS 10a conditions indicate the fastest growth and calcification rates of the record, only comparable to mid-MIS 12 levels, in the beginning of our record, and preceding full glacial conditions. This supports the idea that the coccolithophore community was able to better perform under these transitional conditions, despite colder SST, and under windier and more turbulent settings and more intense upwelling.

Coccolithophore productivity

Coccolithophore productivity

Coccolithophore productivity

Coccolithophore productivity

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evidence from the western Iberian margin for the		
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evidence from the western Iberian margin for the		
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Previous studies in the IbM used coccolith-derived proxies, such as coccolith assemblages, nannofossil accumulation rates (NAR), and total alkenone fluxes (produced mostly by coccolithophores) to reconstruct changes in coccolithophore paleoproductivity (Amore et al., 2012; Maiorano et al., 2015; Marino et al., 2014; Palumbo et al., 2013a). The traditional proxies depend not only on the supply of coccoliths or		

organic compounds but also on dilution by minerals and other sediment constituents and on changing preservation conditions (Rullkötter, 2006). As the CF Sr/Ca ratio is an independent proxy, i.e. independent of sedimentation and accumulation rates, it offers a new perspective into coccolithophore productivity dynamics. We thus assess how well the different productivity proxy records compare with our coccolithophore productivity data.

A visual comparison of our coccolithophore productivity record with the NAR and alkenone flux records already suggests that their relationship is not straightforward (Fig. 8). While in some intervals coccolithophore productivity, NAR and alkenone flux show similar trends, in others, they diverge. Correlation analysis between coccolithophore productivity record and the NAR and the alkenone flux return positive but weak relationships (less than $r=0.3$, statistical analysis not shown).

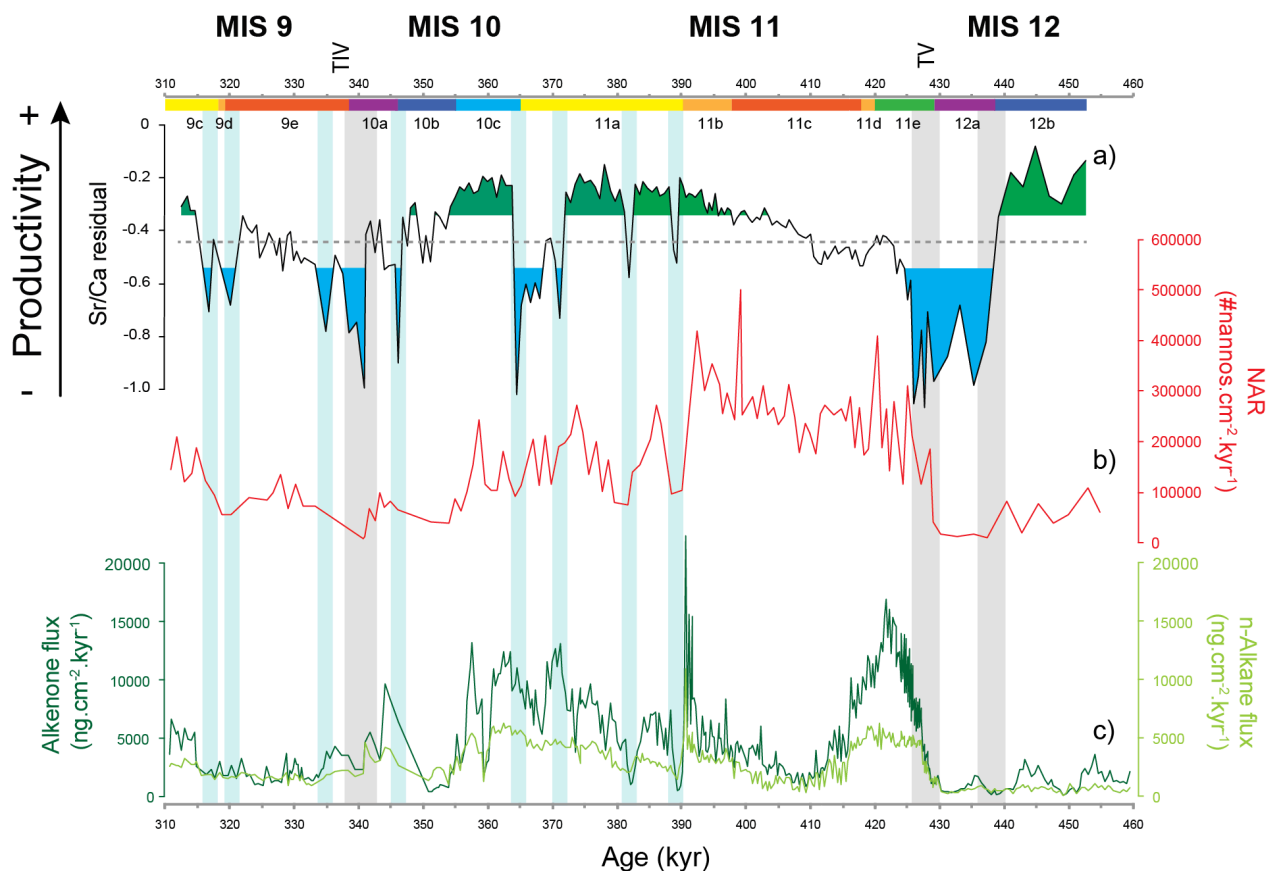


Figure 8 Coccolithophore paleoproductivity reconstruction with “high” and “low” coccolithophore productivity levels highlighted in green and blue shading, respectively (a) compared to: b) NAR (Amore et al., 2012) and c) total alkenone flux and n-alkane flux (Rodrigues et al.,

5.2.1 Coccolithophore productivity and alkenone flux

A synchronous high amplitude increase in the alkenone and the n-alkane fluxes during the deglaciation and beginning of MIS 11 (Fig. 8) made Rodrigues et al. (2011) suggest that coccolithophores had been mostly nourished by a terrestrial input of nutrients either by winds or pluvial discharges. Our coccolithophore productivity record also indicates a synchronous, high amplitude rise. However, in early MIS 11, after the previously mentioned synchronous increase during the deglaciation, the alkenone and n-alkane fluxes show a relatively fast and high amplitude decrease whereas the coccolithophore productivity keeps increasing. Such a mismatch between coccolithophore productivity and alkenone/n-alkane records is also reflected by the very low correlation coefficient ($r=0.08$, $p=0.31$). Moreover, the high positive correlation between the alkenone and n-alkanes fluxes ($r=0.81$, $p<0.01$) suggests that the conditions that allowed a higher continental input, i.e. accumulation and preservation of the n-alkanes (originating from terrestrial plants), also favoured the accumulation and preservation of the alkenone compounds. The discrepancy between our coccolithophore productivity record and the alkenone and n-alkane fluxes suggests that the changes of the latter two mostly reflect fluctuations in the conditions that led to higher export, accumulation and/or preservation of those organic compounds rather than coccolithophore growth, calcification rate and productivity.

We further ran cross-spectral analysis between coccolithophore productivity and alkenone records to check if there were similar semi-periodicities in both time series and respective leads and lags (Fig. 9). In fact, cross-spectral analysis confirms no significant periodicities between the two time series at Milankovitch frequencies. Millennial scale changes are, however, coherent in both time series at ~ 7.8 and ~ 4.4 kyr and the alkenone record leads the coccolithophore productivity by 0.82 and 0.43 kyr, respectively (associated errors of 0.4 and 0.2 kyr, respectively). Coccolithophore productivity leads the alkenone record by 0.9 kyr at quasi-periodicities of ~ 3.7 kyr. Since coccolithophores are the only known organisms producing enough alkenones to get exported and preserved in the sediments in significant amounts, the spectral analysis results corroborate our suggestion that the alkenone record is actually mostly offering a signal of preservation in the sediments.

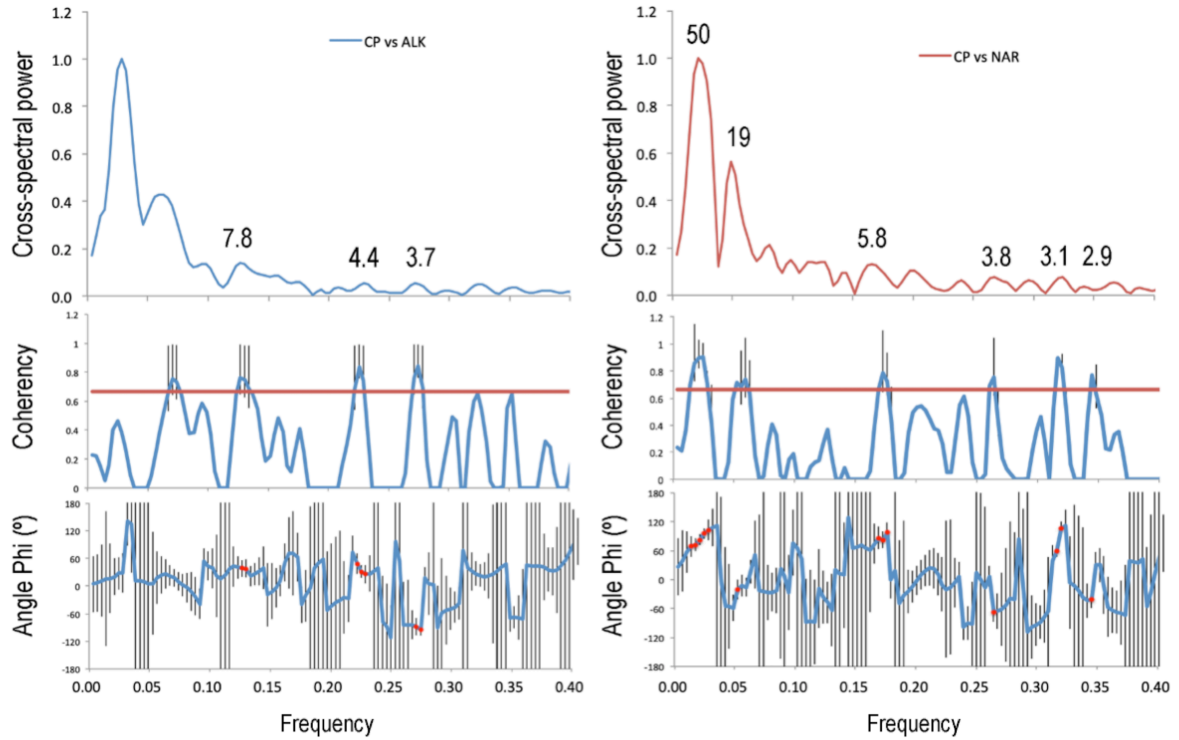


Figure 9 Cross-spectral analysis results for Coccolithophore paleoproductivity (CP) and alkenone flux (ALK – blue line; Rodrigues et al., 2011) and nanofossil accumulation rate (NAR – red line; Amore et al., 2012). The quasi-periodicities are given on top each significant peak on the cross-spectral results. The red line in coherency results defines the significance level of 80% (bandwidth is 0.02). All estimation errors are shown as vertical lines. Red dots highlight the angles for which significant quasi-periodicities exist.

5.2.2 Coccolithophore productivity and NAR

Amore et al. (2012) and Palumbo et al., (2013b) calculated the NAR for core MD03-2699 and suggested generally increased coccolithophore productivity during interglacials compared to glacial substages (Fig. 8). Indeed, both NAR and our coccolithophore productivity record show minima during MIS 12a, MIS 10a and several of the abrupt (HtE) climate events. However, NAR does not reflect increased accumulation during periods of higher coccolithophore productivity, e.g. from 395 kyr to 355 kyr. Contrarily, the NAR reveals large amplitude shifts during times of high coccolithophore productivity.

The coccolithophore productivity and NAR correlation analysis also returned a low correspondence, but higher than with the alkenone flux record ($r=0.28$, $p<0.01$). And, despite coccolithophore productivity and NAR records showing synchronous increasing trends during MIS 11c to MIS 11b, the relationship between the two is not straightforward.

The cross-spectral analysis between the coccolithophore productivity and NAR time series detected that at the millennial time-scales of ~ 5.8 kyr and ~ 3.1 kyr (Fig. 9) NAR is leading coccolithophore productivity by 1.4 kyr and 0.7 kyr, respectively (associated errors of 0.3 and 0.1 kyr, respectively). Since NAR is calculated according to the total number of coccoliths found in the sediments these findings again support

that some specific export or preservation conditions might have happened prior to the increased productivity of coccolithophores. Interestingly, we found coccolithophore productivity leading the NAR at the precession frequency (~ 19 kyr) by 0.1 kyr, as well as at the higher frequencies of 3.8 kyr and 2.9 kyr by 0.72 kyr and 0.33 kyr, respectively. This means that despite some noise within the NAR signal, it does carry our coccolithophore productivity signal, which was not evidenced with the alkenone record.

On glacial to interglacial time scales and during abrupt climate events, the North Atlantic has been subjected to significant changes in the depth of the lysocline (depth below which carbonate starts to dissolve) and in the carbonate compensation depth (depth below which no carbonate is found due to dissolution). We further studied the possibility of dissolution causing the asynchrony between coccolithophore productivity and the NAR. Short-lived dissolution events are likely to have occurred in the deeper IbM area during MIS 12 and MIS 10 glaciations, as suggested by Marino et al. (2014) for core MD01-2446, as a consequence of the northward and upward progression of the southern sourced waters/Antarctic Bottom Water (e.g., Voelker et al., 2010). At a water depth of 1895 m, site MD03-2699 would have been located above the Antarctic Bottom Water influence (placed near 2500 m in the western North Atlantic; Thunell et al., 2002). Two lines of evidence confirm that corrosive southern sourced waters did not affect the coccolithophore assemblages in core MD03-2699: a) there is no evidence for increased abundance of larger, more calcified coccoliths, more resilient to dissolution, in detriment of smaller and less calcified coccoliths in the coccolithophore flora (Amore et al., 2012); b) at IODP Site U1385, located about 500 m deeper than our site, Maiorano et al. (2015) found low coccolith dissolution indices, varying between 1 and 0.9, against the values below 0.25 (MIS 12a) and 0.5 (MIS 10a) for core MD01-2446 at 3570 m (Marino et al., 2014). Since carbonate dissolution does not seem to have been relevant in core MD03-2699, the asynchrony between the NAR and the coccolithophore productivity record must result from other processes.

5.2.3 Why such a mismatch?

One possibility is linked to changes in the phenology of coccolithophore productivity, i.e. changes in the yearly timing when coccolithophores are more productive. Nowadays, coccolithophores in the IbM start to bloom in late winter and spring. Blooms can occur for 6 months (from late winter into summer months) with peak bloom months during spring (April, May; Hopkins et al., 2015). If this phenology was maintained in the past, it would be expected that the coccolithophore productivity record would show a precessional cyclicity, following either spring or summer insolation maxima along with a ~ 6 kyr cyclicity, when productivity would be high at both seasons, such as today, or when a shift to the summer months would occur, most likely associated to colder climatic conditions (e.g., Cavaleiro et al., 2018). Indeed our coccolithophore productivity record shows precession (~ 21 kyr and 17 kyr) as well as ~ 6 kyr quasi-periodicities (harmonics of precession, Fig. 6). This suggests precession, through insolation modulation, as a controlling factor of coccolithophore phenology and thus their productivity. We suggest that coccolithophore phenology in the IbM either remained similar to today or shifted to a predominant summer

productive peak during cold substages and stadials, and as already suggested to the mid North Atlantic (Cavaleiro et al., 2018).

To check for the relationship between coccolithophore productivity and insolation at precession and precession harmonics we ran cross-spectral analysis with equinoxes and solstices peak daily insolation (Fig. 10). The results returned a non-significant peak around 6 kyr for spring, whereas summer solstice insolation led coccolithophore productivity by 1.5 kyr (± 0.34 kyr) and autumn equinox and winter solstice lagged coccolithophore productivity by 0.9 kyr (± 0.36 kyr) and 1.7 kyr (± 0.28 kyr), respectively. This corroborates the theory that coccolithophore phenology might have changed from spring to summer during colder periods while autumn and winter insolation changes had a diminished effect on coccolithophore productivity.

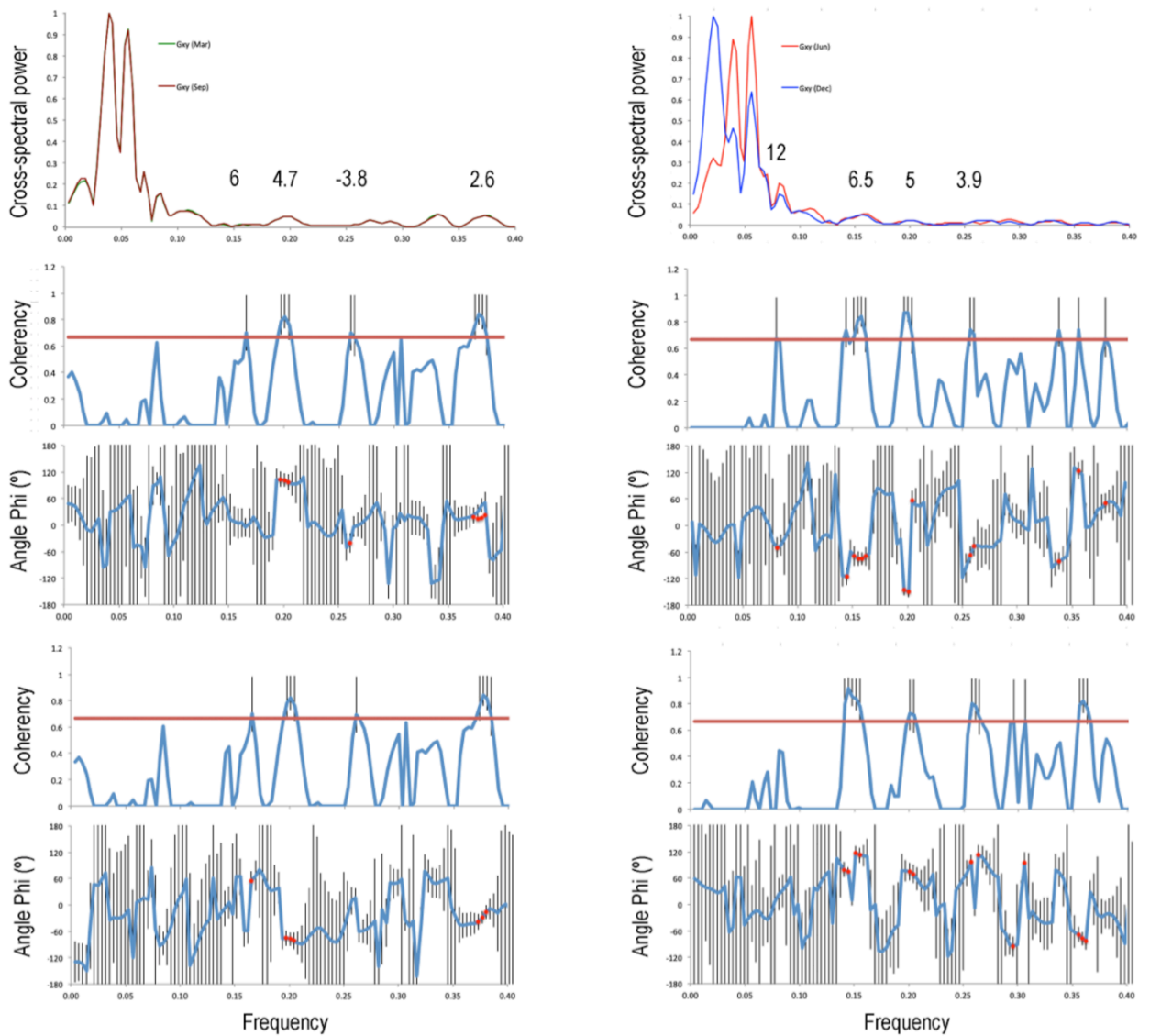


Figure 10 Cross-spectral analysis results for Coccolithophore paleoproductivity (PC) and insolation curves: March and September cross spectral analysis results (left panel), June and December cross spectral analysis

results (right panel; insolation data from Laskar et al., 2011). The quasi-periodicities are given on top the first four significant peaks on the cross-spectral results. The red line in coherency results defines the significance level of 80% (bandwidth is 0.02). All estimation errors are shown as vertical lines. Red dots highlight the angles for which significant quasi-periodicities exist.

If such changes in phenology did happen, then, during periods of colder climate in the IbM, coccolithophores would be forced into a narrower time window to flourish (i.e. increased insolation, higher nutrient availability and less competition with diatoms). Coccolithophore productivity would thus rise when nutrients and insolation increase but the yearly time-window for higher productivity would become narrower as SST would decrease as a consequence of the more frequent arrival of (sub)polar waters. This could ultimately lead to lower NAR and decreased accumulation of alkenones as well, even at times when coccolithophores were calcifying faster and more efficiently because the yearly time window for export to happen would be narrower.

6 Conclusions

With this study we discuss the different factors controlling coccolithophore productivity in the IbM. Coccolithophore productivity is primarily affected by climatic changes, namely changes in temperature and ocean circulation that led to the predominance of different water masses in the IbM. Presence of (sub)polar surface water masses during glacial substages and stadials resulted in lower coccolithophore productivity, in particular during HtE stadials. On the other hand, the prevalence of nutrient-poorer subtropical waters during interglacial substages and increased riverine input of iron and silica increased coccolithophores' and diatoms' competition for nutrients leading to relative intermediate coccolithophore productivity levels. The transitions between interglacial and glacial substages were characterized by coccolithophore productivity maxima due to the emerging presence of nutrient-rich waters brought by strong northerly wind forced upwelling. These factors allowed coccolithophores to bloom and attain their relative calcification rate maxima and ultimately their productivity maxima. During the transition from interglacial to glacial substages coccolithophores were nonetheless forced to change their phenology, and their most productive season changed according to the best conditions provided, most likely from spring to summer because the SSTs during winter and spring were still too cold. Ultimately, this contributed to the dissonance between coccolithophore productivity, NAR and alkenone flux records, as a consequence of the yearly time-window narrowing of coccolithophore productivity.

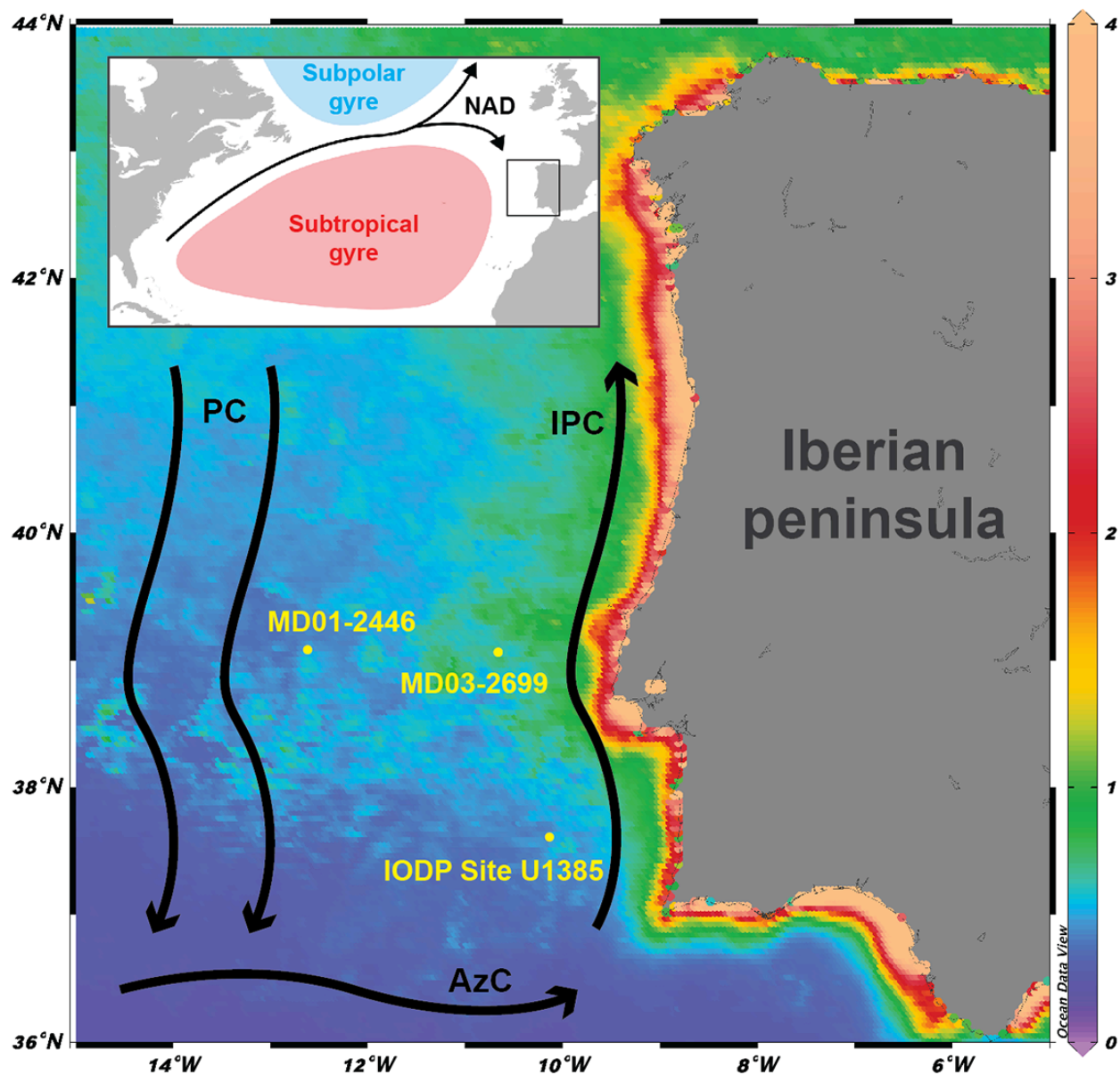


Figure 1: Core location and major currents in the area: NAD = North Atlantic Drift; PC = Portugal Current; IPC = Iberian Poleward Current; AzC = Azores Current. Background: chlorophyll *a* concentration (mg.m⁻³; March, April and May average, 2003-2018) derived from MODISA satellite data available at <http://disc.sci.gsfc.nasa.gov/giovanni>.

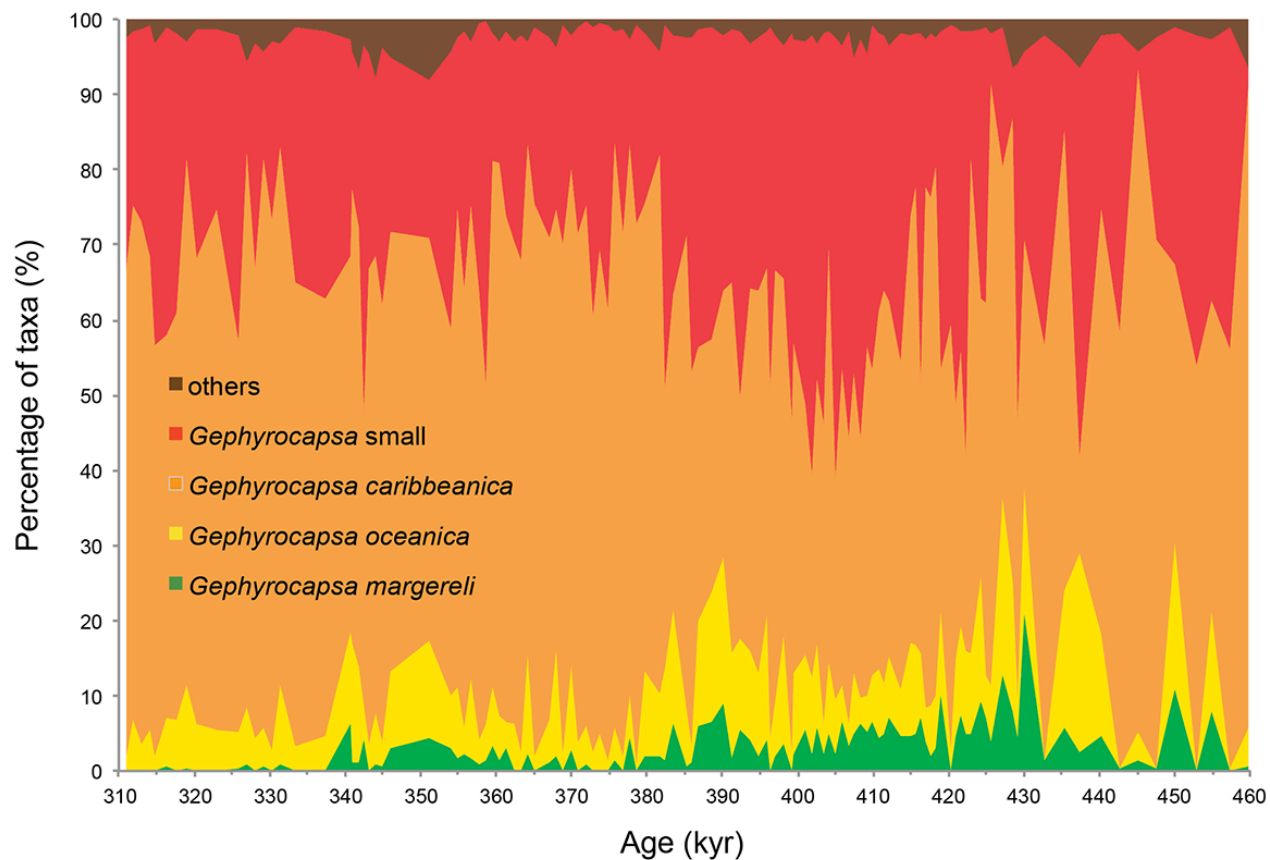


Figure 2: Composition of coccolith assemblages in core MD03-2699 based on Amore et al. (2012). Note that only a minor percentage of coccoliths belong to groups other than the dominant Gephyrocapsids. The chronology is based on the age model of Voelker et al. (2010).

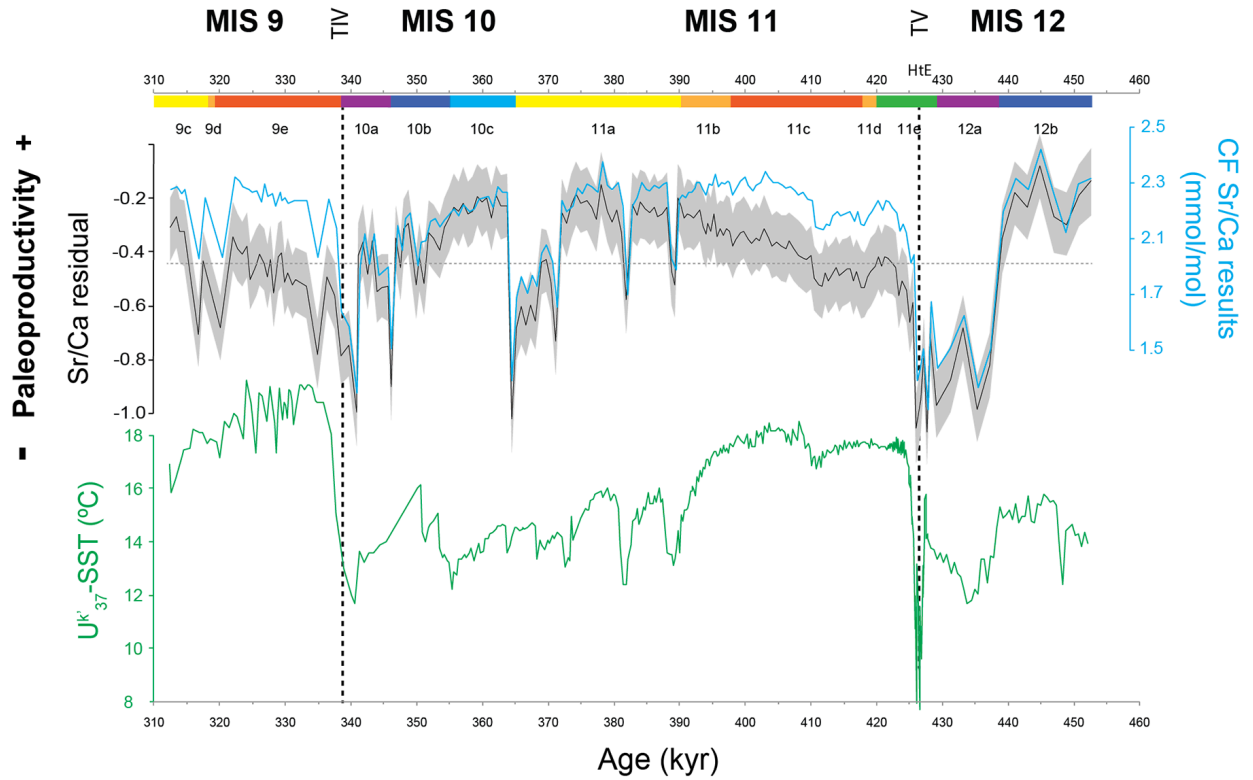


Figure 3: Coccolithophore paleoproductivity results from core MD03-2699: coccolith fraction (CF) Sr/Ca ratio and the resulting CF Sr/Ca residual with confidence interval (grey shading represents Monte Carlo 20-80% interval), U^k₃₇-based reconstruction of sea surface temperature (Rodrigues et al., 2011) The horizontal dashed line marks the average value for CF Sr/Ca and CF Sr/Ca residual. The chronology is based on the age model of Voelker et al. (2010), Marine isotope stages (MIS) and substages marked according to Railsback et al. (2015), vertical dashed lines highlight Terminations V and IV.

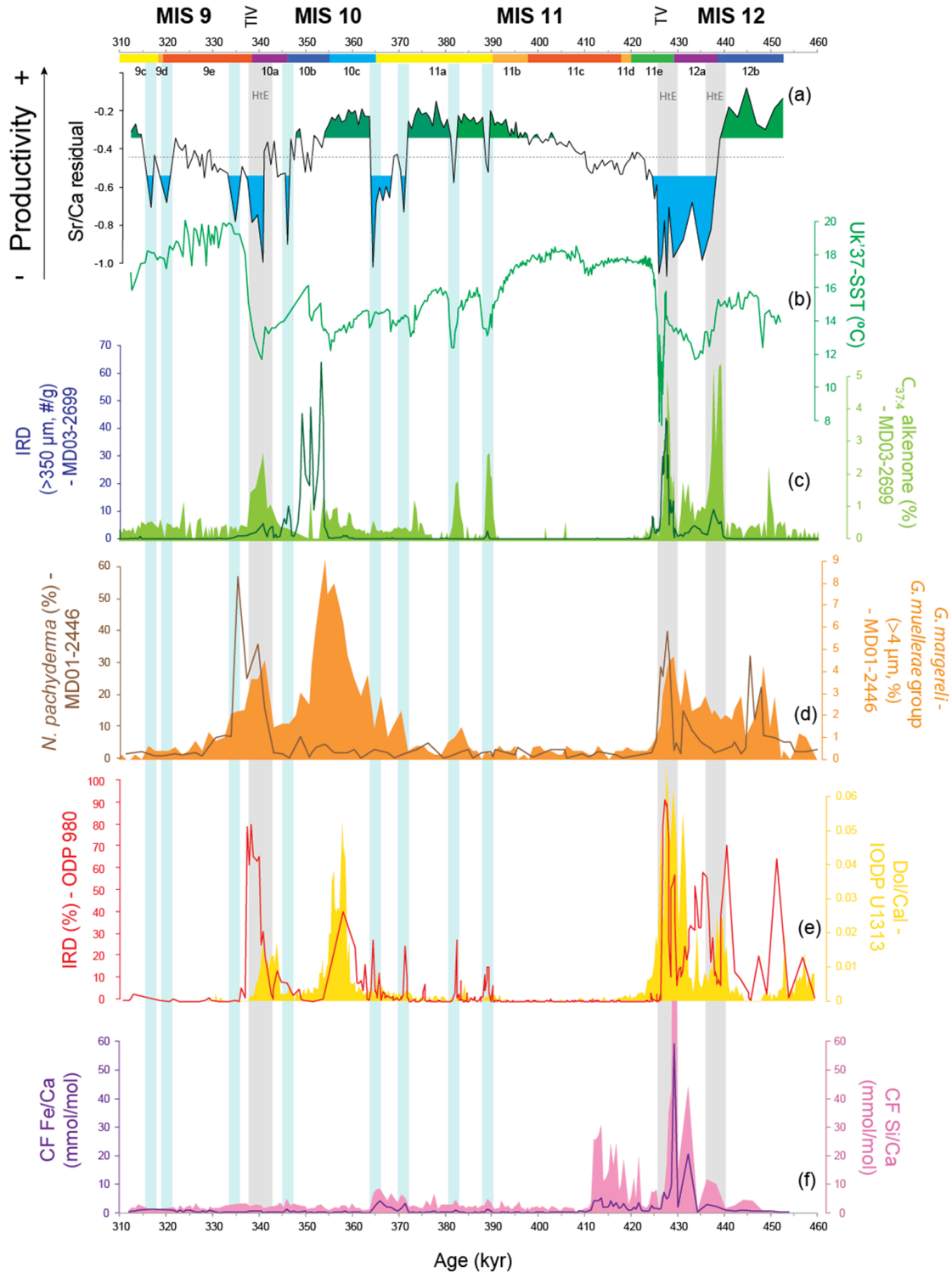


Figure 4: Coccolithophore paleoproductivity reconstruction with “high” and “low” coccolithophore productivity levels highlighted in green and blue shading, respectively (a) compared to: b) U^k₃₇-based reconstruction of sea surface temperature from core MD03-2699 (Rodrigues et al., 2011); c) ice-rafted debris (IRD) abundance (Voelker et al., 2010) and heptatriatetraenone (C_{37:4} alkenone) abundance (Rodrigues et al., 2011), both from core MD03-2699; d) *Neogloboquadrina pachyderma* relative abundance and *Gephyrocapsa margereli* and *G. muelleriae* group relative abundance, both

from core MD01-2446 (Marino et al., 2014); e) percentage ice-rafted debris (IRD) from ODP Site 980 (McManus et al., 1999) and Dolomite/Calcite ratio from IODP Site U1313 (Stein et al., 2009); and f) coccolith fraction (CF) Fe/Ca and Si/Ca ratios. Note that the MD03-2699 lithic fragments were counted in a coarser size fraction ($>315\ \mu\text{m}$) than the standard size fraction ($>150\ \mu\text{m}$; Hemming et al., 2004) and thus most likely only record major ice-rafting events at the IbM, as suggested by Marino et al. (2014). Chronology as in Fig. 3. Vertical bars: grey bars correspond to Heinrich-type events and blue bars to short-lived events of decreased coccolithophore productivity.

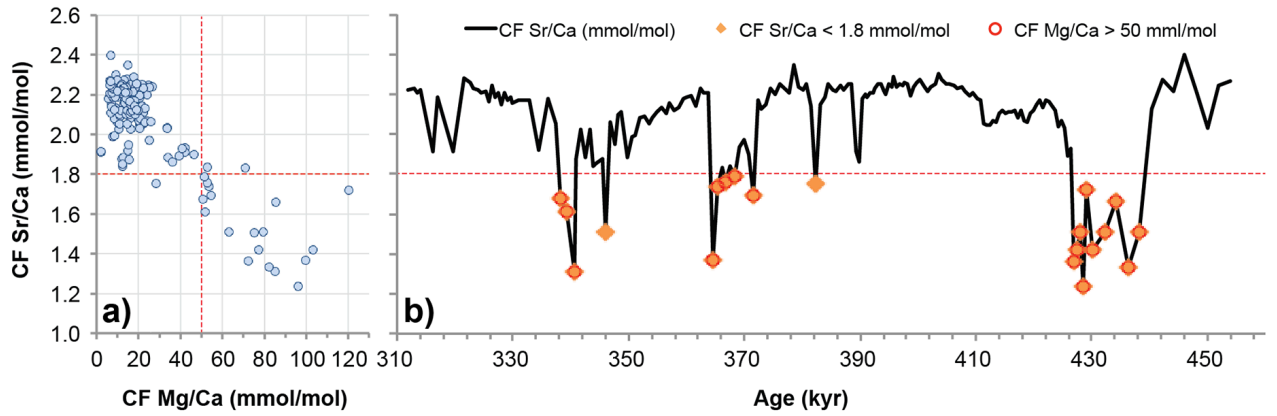


Figure 5: Detrital carbonate analysis: a) coccolith fraction (CF) Sr/Ca results and CF Mg/Ca cross-plot. Vertical and horizontal red lines delimit thresholds at 1.8 mol/mol of Sr/Ca and 50 mmol/mol of Mg/Ca, respectively; b) CF Sr/Ca samples possibly subjected to detrital carbonate contamination (red makers), notably associated with cold and abrupt events.

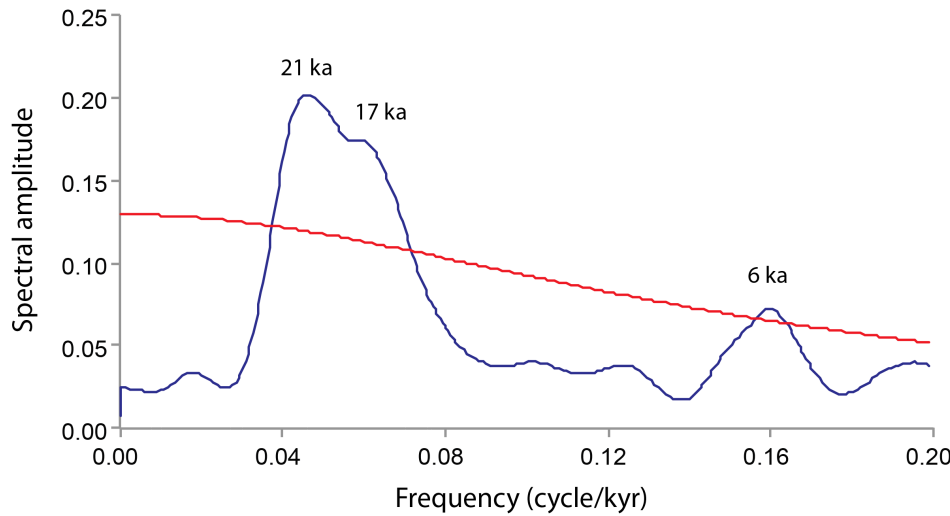
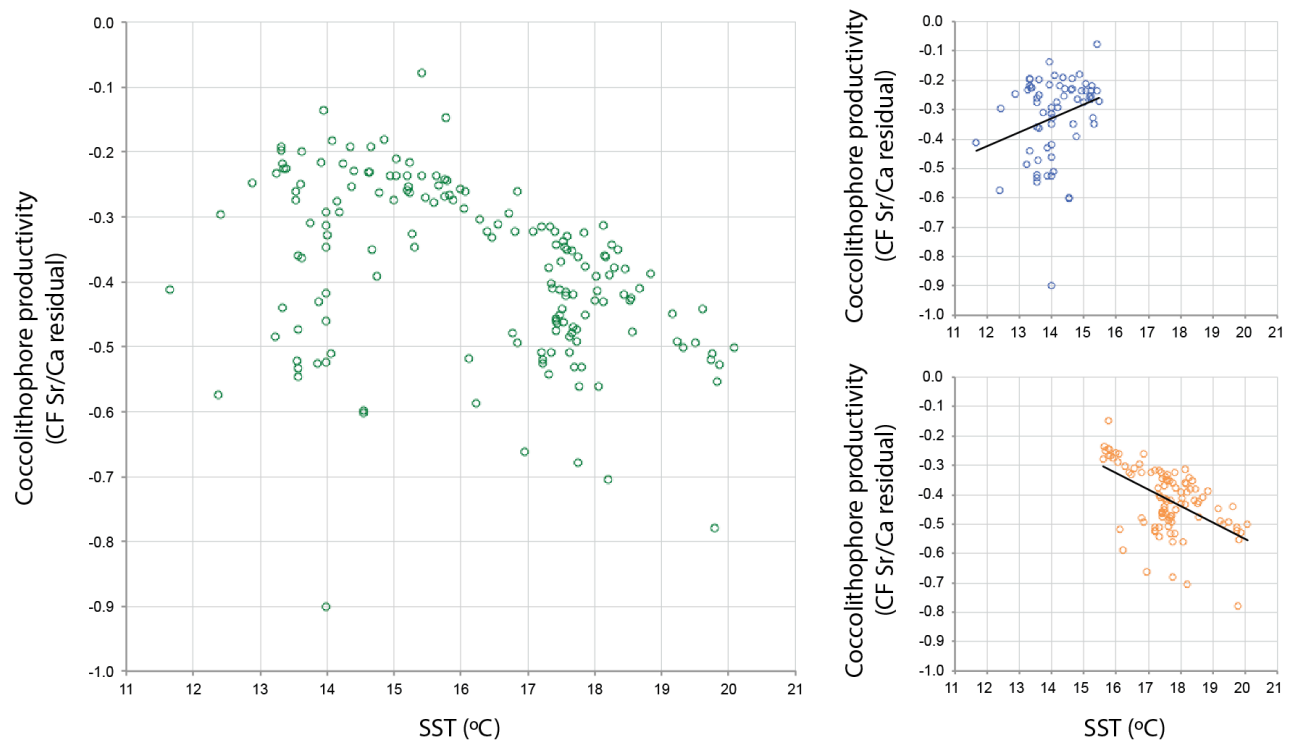


Figure 6: Coccolithophore paleoproductivity time-series analysis showing significant peaks at 21, 17 and 6 ka. The red line defines the significance level of 80% (bandwidth is 0.02), indicating that the peaks at 21 and 17 ka are not independent (may represent the same frequency given the uncertainty of dating and resolution).



Correlation Coefficients - Coccolithophore productivity (CF Sr/Ca residual) vs SST (°C)

	<i>Sr/Ca residual vs. SST (°C)</i>	<i>Sr/Ca residual vs. SST (°C)</i>	<i>Sr/Ca residual vs. SST (°C)</i>
R	-0.4	0.27	-0.54
<i>R Standard Error</i>	<i>5.17E-3</i>	<i>0.01</i>	<i>7.3E-3</i>
<i>t</i>	<i>-5.52</i>	<i>2.22</i>	<i>-6.32</i>
<i>p-value</i>	<i>1.29E-7</i>	<i>0.03</i>	<i>7.8E-9</i>
<i>H0 (5%)</i>	<i>rejected</i>	<i>rejected</i>	<i>rejected</i>
<i>No# of valid cases</i>	<i>165 (all valid samples)</i>	<i>66 (samples SST < 16 °C)</i>	<i>99 (samples > 16 °C)</i>

Figure 7: Coccolithophore paleoproductivity and SST Pearson correlation results: all valid samples (green), samples below 15.5 °C (blue) and samples above 15.5 °C (orange). Person correlation computed with StatPlus:mac, AnalystSoft Inc. - statistical analysis program for macOS®. Version v7. See <http://www.analystsoft.com/en/>

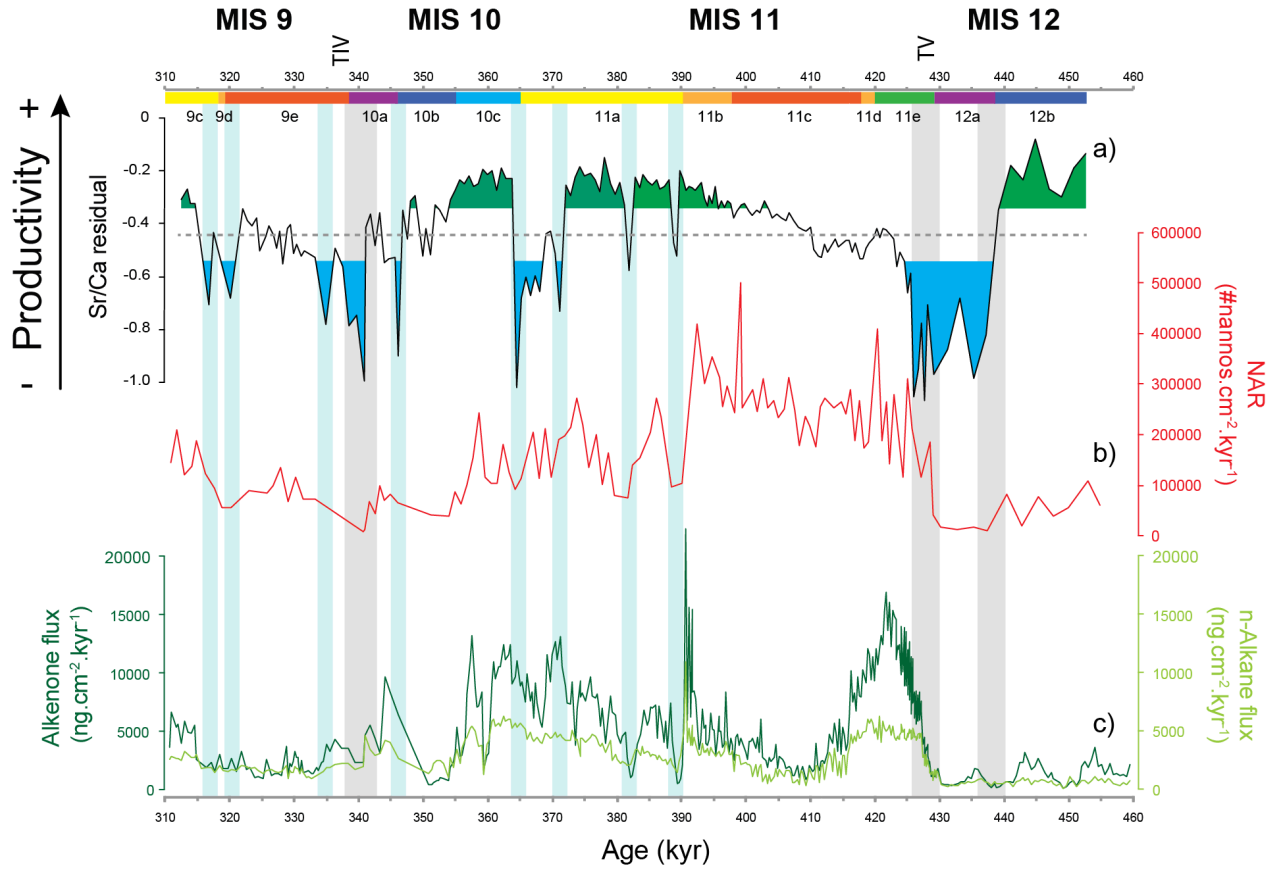


Figure 8: Coccolithophore paleoproductivity reconstruction with “high” and “low” coccolithophore productivity levels highlighted in green and blue shading, respectively (a) compared to: b) NAR (Amore et al., 2012) and c) total alkenone flux and n-alkane flux (Rodrigues et al., 2011). Chronology as in Fig. 3. Vertical bars: grey bars correspond to Heinrich-type events and blue bars to short-lived events of decreased coccolithophore productivity.

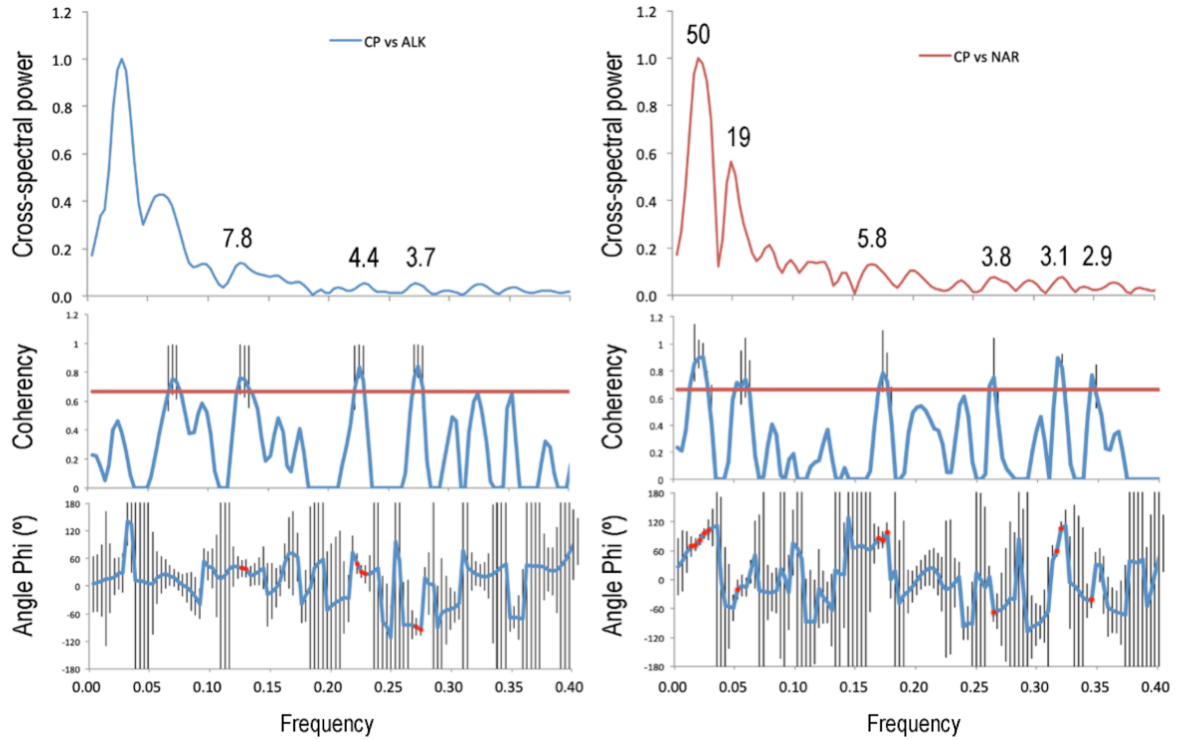


Figure 9: Cross-spectral analysis results for Coccolithophore paleoproductivity (CP) and alkenone flux (ALK – blue line; Rodrigues et al., 2011) and nannofossil accumulation rate (NAR – red line; Amore et al., 2012). The quasi-periodicities are given on top each significant peak on the cross-spectral results. The red line in coherency results defines the significance level of 80% (bandwidth is 0.02). All estimation errors are shown as vertical lines. Red dots highlight the angles for which significant quasi-periodicities exist.

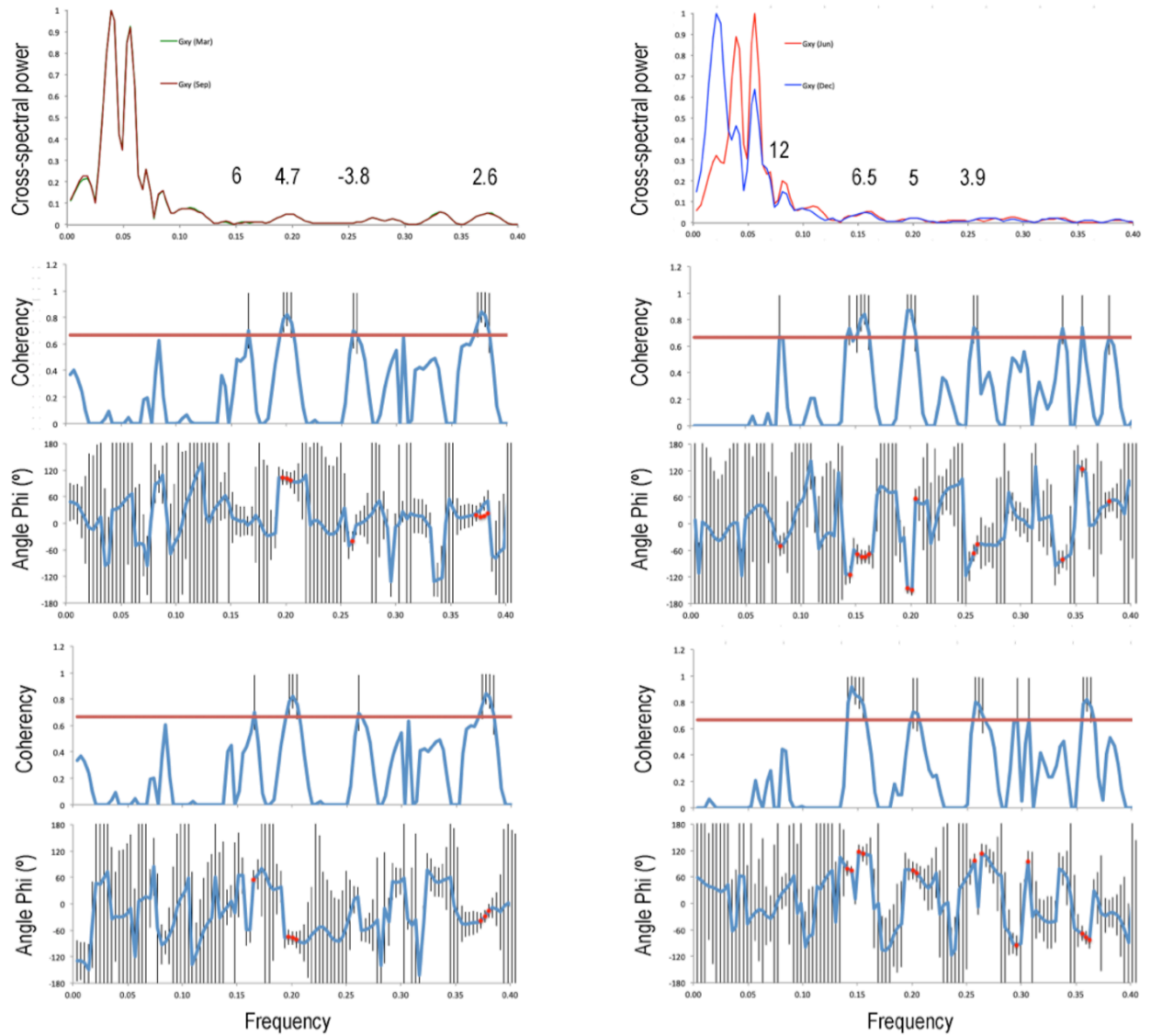


Figure 10: Cross-spectral analysis results for Coccolithophore paleoproductivity (PC) and insolation curves: March and September cross spectral analysis results (left panel), June and December cross spectral analysis results (right panel; insolation data from Laskar et al., 2011). The quasi-periodicities are given on top the first four significant peaks on the cross-spectral results. The red line in coherency results defines the significance level of 80% (bandwidth is 0.02). All estimation errors are shown as vertical lines. Red dots highlight the angles for which significant quasi-periodicities exist.