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**Multi-species
sedimentary
coccolith
calcification analysis**

P. R. Halloran et al.

A multi-species coccolith volume response to an anthropogenically-modified ocean

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Abstract

Major questions surround the species-specific nature of coccolithophore calcification in response to rising atmospheric CO₂ levels. Here we present CaCO₃ particle volume distribution data from the coccolith size-fraction of a rapidly accumulating North Atlantic sediment core. These data appear to indicate that coccoliths produced by the larger coccolithophore species present at this location increase in mass in parallel with anthropogenic CO₂ release. This finding has significant implications for the realistic representation of an assemblage-wide coccolithophore CO₂-calcification response in numerical models.

1 Introduction

Phytoplankton mineralization in undersaturated waters is a widespread feature of the oceans, as evidenced by thriving diatom and acantharia populations in waters undersaturated in silica and strontium sulphate respectively. Recent laboratory studies indicate that physiological controls over mineralization, in some phytoplankton, can overcome reductions in the saturation state of ambient seawater (Iglesias-Rodriguez and Halloran et al., 2008; Langer et al., 2006a; Langer et al., 2006b). Furthermore, *Emiliania huxleyi* has demonstrated an ability to calcify at a saturation state of 0.4 (Henderiks and Rickaby, 2007). Coccolithophores calcify intracellularly, and as such, the complexity of their calcification response to a modified external seawater carbonate system may result from a variable biological uptake of different forms of dissolved inorganic carbon for calcification, or differences between the manipulation of culture experiments, but may also reflect the physiological tolerance and carbon concentration ability of different coccolithophores (Iglesias-Rodriguez and Halloran et al., 2008).

An increase in the average coccolith mass, independent of a change in species composition, has been demonstrated to parallel the rise in atmospheric $p\text{CO}_2$ over the past ~220 years, by analysis of the <10 μm sediment fraction from a Sub-polar North

BGD

5, 2923–2930, 2008

Multi-species sedimentary coccolith calcification analysis

P. R. Halloran et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Atlantic core (RAPID 21-12-B, 57°27.09'N, 27°54.53'W, situated at 2630 m) (Iglesias-Rodriguez and Halloran et al., 2008). Here we present a size-distribution analysis of the RAPID 21-12-B <10 μm size fraction, indicating that the increase in coccolith mass occurs across a broad range of coccolithophore species.

2 Methodology

Sediment core RAPID 21-12-B was sampled at 0.5 cm intervals. Approximately 2 to 4 g of the total sediment from each sample was suspended in buffered H_2O and passed through a 10 μm pore-size track-etched polycarbonate filter membrane, using bubble agitation to prevent blocking. Three sub-samples were taken from the <10 μm fraction, each suspended in 100 ml of electrolyte (ISOTON II), and the particle volume frequency distribution of each fraction, between 0.63 and 10 μm equivalent spherical diameter (ESD), analysed three times using a Beckman Coulter Multisizer III Coulter Counter fitted with a 30 μm aperture. This technique provides a direct measure of particle volume, by recording the volume of electrolyte displaced as the particle passes through the sensing zone of the aperture. These data were collected in 256 bins distributed on a log basis across the 9.37 μm sizing window. After replicate analysis, 500 μl of concentrated HCl was added to each suspension, stirred, left to stand, then stirred again vigorously for 4 min before re-analyzing each sub-sample three times. We have then calculated the difference between pre-acidification and post-acidification volume frequency distributions (typically totaling 80 000 CaCO_3 particles). In order to allow direct comparison of CaCO_3 volume frequency distribution across the time-series, the average of the nine measured replicate CaCO_3 particle counts in each of the 256 bins has been divided by the average of the nine measured replicate total CaCO_3 particle counts in that sample. We consider these data to characterize the <10 μm sedimentary CaCO_3 particle distribution, representing the coccolith fraction, comprising *Emiliania huxleyi*, small *Gephyrocapsa*, *Gephyrocapsa oceanica*, *Gephyrocapsa muelleriae*, *Gephyrocapsa caribbeanica*, *Oolithotus fragilis*, *Calcidicus lept-*

BGD

5, 2923–2930, 2008

Multi-species sedimentary coccolith calcification analysis

P. R. Halloran et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

porus, *Coccolithus pelagicus* var. *pelagicus*, *Helicosphaera carteri* (Iglesias-Rodriguez and Halloran et al., 2008). The high abundance of small coccoliths in the RAPID 21-12-B sediments makes it necessary to normalize the particles counts in each bin by the average number of counts in all bins of that volume. This data is presented as a percentage anomaly from that average value, and allows a clearer comparison of trends occurring in different volume ranges.

3 Results and discussion

The average coccolith mass increases by ~40% over the last 220 years (Fig. 3 in Iglesias-Rodriguez and Halloran et al., 2008), and this shift is associated with a relative increase in the frequency of the large CaCO_3 particles ($>2\mu\text{m}$ ESD) compared to the small CaCO_3 particles ($<2\mu\text{m}$ ESD) (Fig. 1, Supplementary materials <http://www.biogeosciences-discuss.net/5/2923/2008/bgd-5-2923-2008-supplement.zip> dataset, and Supplementary materials Table 1). Given that the coccolith species composition is invariant over this interval (2005), the trend towards larger CaCO_3 particles can be interpreted as an increase in the volume of coccoliths produced by a range of coccolithophore species. This increase in coccolith volume spans the observed volume range of coccoliths produced by the larger coccolithophore species present in the North Atlantic (Young and Ziveri, 2000). If the increase in average coccolith mass was driven by a change in calcification of a single species, the size-distribution data would show decreasing particle counts in the volume-region initially occupied by that species, coincident with an increasing CaCO_3 particle frequency in the larger volume-region. Comparison of the red and black lines in Fig. 1b (representing the size distribution over the last 12 years, and the pre-industrial to 1960 AD averaged signals respectively), shows that the coccolith mass increase occurs across a wide range of the species typically larger than $2\mu\text{m}$ ESD.

Our relative increase in the frequency of large particles is necessarily accompanied by a relative decrease in the frequency of small particles. In order to allow examination

**Multi-species
sedimentary
coccolith
calcification analysis**

P. R. Halloran et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

of the subtle changes occurring within the frequency of the abundant small particles, Fig. 2 presents the volume frequency anomalies in log space. The negative frequency anomaly between 1 to 4 μm ESD, encompassing the upper and lower end member size ranges of the smaller and larger coccolithophores respectively, which develops towards the core-top suggest that coccoliths are moving away from this volume range. This reaffirms that the volume of the coccoliths produced by the larger coccolithophores (*O. fragilis*, *C. leptoporus*, *C. pelagicus* var. *pelagicus*, *H. carteri*) has increased. In addition, the recent relative increase in particle frequency below 1 μm ESD suggests either that, coccoliths produced by the smaller coccolithophores (*E. huxleyi*, *G. oceanica*, *G. muelleriae*), are becoming more lightly calcified, or that the decreasing pH of the modern ocean is promoting partial dissolution of these delicate coccoliths. This apparent recent divergence between the volumes of these two size-groups could reflect contrasting physiological controls and evolutionary adaptation.

4 Conclusions

In order to reduce the uncertainty associated with the calcification CO_2 feedback within Earth system models, we need to fully constrain and understand the inter-specific variability of coccolithophore calcification to conditions of ocean acidification. Our sedimentary data appear to indicate that in the real ocean the larger coccolithophore species increase their calcification in response to anthropogenic CO_2 release. If the change in coccolith size distribution presented here is representative of that elsewhere in the ocean, it may be possible to describe the coccolithophore calcification response to rising levels of atmospheric CO_2 at an assemblage level. Such a description could be incorporated into carbon-cycle models without the need to parameterize individual species, and therefore avoid uncertainty resulting from our poor understanding of the complex inter-species variability.

The next step towards a full understanding of the coccolithophore calcification response over the industrialized era, will be to combine individual species size analysis,

**Multi-species
sedimentary
coccolith
calcification analysis**

P. R. Halloran et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

taking advantage of new techniques such as that presented by Beaufort (2000), with Coulter Counter analyses capable of measuring coccolith volumes, to constrain how calcification has changed at a species level. Experiments can then be designed and applied to new high-resolution cores, extending the observations we have made to a range of oceanic regimes.

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BGD

5, 2923–2930, 2008

Multi-species sedimentary coccolith calcification analysis

P. R. Halloran et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Multi-species sedimentary coccolith calcification analysis

P. R. Halloran et al.

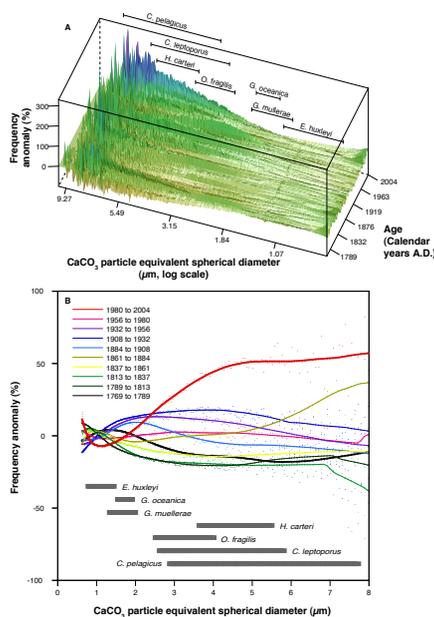


Fig. 1. (a) 3-D surface showing the percentage anomaly in frequency data (with respect to the average value for each volume-bin) for CaCO_3 particles (coccoliths) measured in samples from 1770 to 2004 AD. Black bars above the main portion of the figure represent the equivalent spherical diameter size-ranges occupied by coccoliths of the major species present in these samples, calculated from Young and Ziveri (2000). (b) Curves representing the size-distribution of particles averaged over ~24 year (5.5 cm down core) intervals, presented as a percentage anomaly from the average frequency for each size-bin. Heavy red and black lines represent the most recent, and oldest, data respectively. Data has been fitted with a 15% least squares weighted mean. Data has been truncated at $8 \mu\text{m}$ because particle counts in bins larger than $8 \mu\text{m}$ are low, and therefore when ratios are formed data becomes noisy, detracting from the useful signal. Figure 3 presents the full dataset, negating this issue. Grey bars represent the typical range of coccolith-volume equivalent spherical diameters, Fig. 2. Contour plot representing the evolution of the particle volume-distribution in log-space, to emphasize the subtle percentage change in the abundant small-volume particles. Colors represent log values of the percentage anomaly in frequency data (with respect to the average value for each size-bin) for CaCO_3 particles (coccoliths) measured in samples spanning 1770 to 2004. Note that negative anomalies are represented by -1 multiplied by the log of the modulus of the percentage frequency anomaly.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

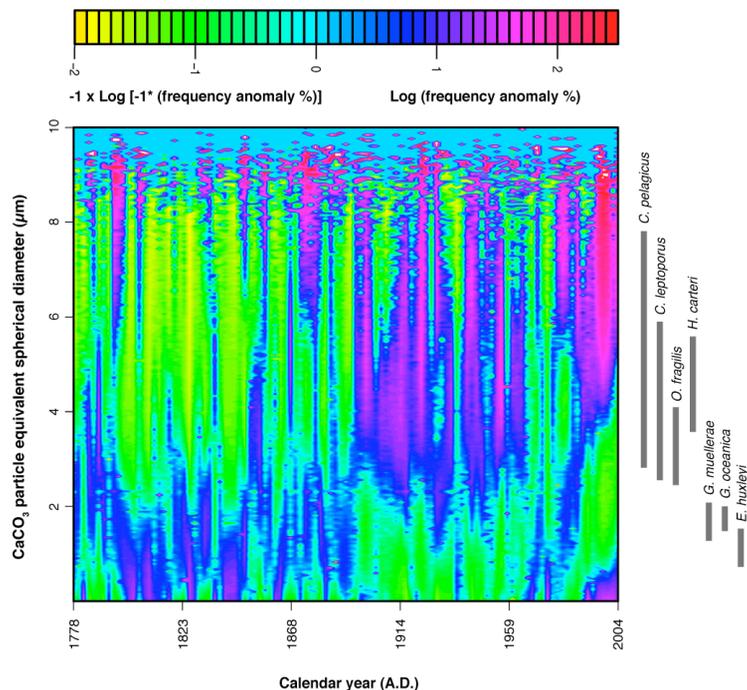


Fig. 2. Contour plot representing the evolution of the particle volume-distribution in log-space, to emphasize the subtle percentage change in the abundant small-volume particles. Colors represent log values of the percentage anomaly in frequency data (with respect to the average value for each size-bin) for CaCO_3 particles (coccoliths) measured in samples spanning 1770 to 2004. Note that negative anomalies are represented by -1 multiplied by the log of the modulus of the percentage frequency anomaly

Multi-species sedimentary coccolith calcification analysis

P. R. Halloran et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion