

## ***Interactive comment on “Environmental controls on the *Emiliana huxleyi* calcite mass” by M. T. Horigome et al.***

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We wish to thank Poulton and colleagues for the detailed analysis of our paper which have been very helpful and greatly improved the quality of this manuscript. A detailed reply to each comment follows below:

1) What evidence exists that these are Holocene sediments? The assertion that the sediments used in this study are ‘pre-industrial/late Holocene/modern age’ is fundamental to the conclusions and methods applied. However, no age-models or dating of the sediment is presented in the paper, only a reference to a book chapter by one of the co-authors (Baumann et al. 2004). When looking at this book chapter, the methods only state: “All sediments are assumed to be of Holocene age. However, only a few of these surface sediments have been dated, nor is the exact sedimentation rate known at

Full Screen / Esc

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most sites. The ages of surface sediments may vary from decades to several hundreds, or even up to several thousands of years, depending on the local sedimentation rate.” This highlights several issues in terms of the age of the sediments used in the paper by Horigome et al., such as the assumption that the sediments are pre-industrial in origin. How confident can the authors be that these are pre-industrial Holocene sediments? How do they know what proportion of the sediment is post-industrial vs. pre-industrial? This is a key potential weakness of the study, that without being addressed may undermine the entire work – when were these coccoliths deposited? Also, things in the ocean do not sink straight down, there is considerable lateral movement in the upper ocean. Hence, is it suitable to correlate point X on the sea floor with a source region directly above it, especially accounting for the strong oceanic gradients of the study areas – what evidence do the authors have for little or no lateral movement of their coccoliths? Additionally, bioturbation of the surface sediment by burrowing organisms is likely to have mixed the top sediment to varying degrees at different sites. This also affects the age of the coccoliths, much older sediment could potentially be mixed up to the surface.

Author Response: One of our prime assumptions is that the age of the studied sediments range between modern (pre-industrial) and Late Holocene, thus, making the generated sedimentary record directly comparable with pre-industrial surface ocean physicochemical properties. We appreciate the remarks of Poulton and colleagues that prompted us to realize our rather informal treatment of this issue. However, we are convinced of pre-industrial age for all of the investigated material, despite not having direct age control on most of the samples analysed. For those samples in which  $^{14}\text{C}$  and/or  $^{210}\text{Pb}$  data are available (i.e., 1413-2, 1414-2, 1415-1, 1417-1, CD154-01-01K, CD154-02-03K, CD154-03-05K, CD154-05-07K, CD154-10-10K, and MD02-2594) modern to late Holocene ages are confirmed (Mollenhauer et al., 2004; Martinez-Mendez et al., 2010; Negre et al., 2010; Jonkers et al., 2012). In addition, the basic evidence that the remaining samples from the wider South Atlantic region are at least of Late Holocene age (and not older) comes from a couple of (nearby)

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dated core-tops in the South Atlantic (Mollenhauer, 2002, Mollenhauer et al., 2003, 2004, 2006, 2007), and from a number of investigated sediment cores from the entire study region (e.g., various articles in Wefer et al., 2004). The available core-top radiocarbon ages display a large variability ranging mostly between modern and about 3500  $^{14}\text{C}$  years (e.g., 2730 – 3470  $^{14}\text{C}$  years for GeoB 1112-3; Mollenhauer, 2002). A few “old” core top dates up to 8000 radiocarbon years (Mollenhauer et al., 2006) are from piston cores and probably reflect the loss of sediment during the coring process. Despite the relatively large variability in age, the available data collectively rule out the possibility that our dataset is influenced by glacial sediments (and coccoliths), which calcified under physicochemical conditions in both surface- and deep-ocean that were substantially different from the modern/Holocene (e.g., Hönisch & Hemming, 2005; Foster, 2008; Yu et al., 2010). In Table 1 of the revised manuscript, we will provide the sedimentation rates (from the nearest radiometrically dated records, or from the same sites considered in this study) from previously published studies.

We also appreciate the concern of Poulton and colleagues about potential contamination by post-industrial material. We stress, however, that ages containing bomb-radiocarbon (i.e., younger than 1950s), in contrast, were only observed in the South Atlantic in a few areas with extremely high sedimentation rates (up to  $> 20$  cm/kyr; see compilation in Baumann et al., 2004, Mollenhauer et al., 2004). Since those areas are limited to the uppermost continental margins and shelf areas (see Fig. 4 in Mollenhauer et al., 2004), we expressly decided not to sample these sectors of the South Atlantic for our study. Hence we are confident that we only chose pre-industrial Holocene sediments. Sedimentation rates in the areas selected for the present study are between about 1 cm/kyr up to about 4-6 cm/kyr, which de facto rule out a verifiable influence of post-industrial sediments.

Vertical displacement by bioturbation has been extensively discussed in the literature (e.g., Trauth et al., 1997, Thomson et al., 2000, Anderson et al., 2001, Bard, 2001). If bioturbation is assumed to create a homogenized mixed layer in the sediment, the

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potential effect on core-top radiocarbon ages can be estimated (as has been done by Mollenhauer et al., 2007): The radiocarbon age of a homogenized layer of 10 cm thickness would be approximately 5970 14C years, if the sedimentation rate were 1 cm kyr<sup>-1</sup>. At sedimentation rates of 3, or 5 cm kyr<sup>-1</sup>, the age of the 10 cm-thick homogenized mixed layer would be 2650, or 1940 14C years, respectively. The available core-top ages up to 3500 14C years fall within the range of calculated values for sedimentation rates between 1 and 3 cm kyr<sup>-1</sup> and would thus imply that these “old” core-top ages were the result of homogenization of the upper 10 cm of the sediment. However, these are maximum ages, indicating that bioturbation is not mixing up “much older” (i.e., glacial) sediments to the sediment-water interface.

We acknowledge that the sinking of particles is not vertically downward, because of the ocean currents. However, the flux of materials to the deep-sea is dominated by larger, organic-rich particles with sinking rates up to several hundred meters per day (e.g. Fischer & Karakas, 2009). Fecal pellets are thought to be the major carrier for coccolithophores to the deep ocean (e.g., de La Rocha and Passow, 2007). Such particles containing high amounts of coccoliths can reach sinking rates of up to several hundreds meters per day (Ploug et al., 2008). Therefore it is not surprising that comparisons of coccolithophore plankton communities with those of underlying sediments (Baumann et al., 2000, Boeckel & Baumann, 2008) have shown that the distribution of the water column coccolithophores is well reflected in the sedimentary archive, thus proving their potential as paleoecological archive/proxy systems. Hence, we conclude that it is suitable to that point X on the sea floor either correlates with a source region directly above it (if no lateral displacement is considered) or integrates the “information” over a slightly larger area of the surface ocean (if small lateral displacement is considered).

2) The paper describes changes in coccolith size, not mass. The calcite content of *Emiliania huxleyi* coccoliths is directly proportional to their size (see Young and Ziveri 2000 Deep-Sea Research II). Hence this manuscript shows a clear change in coccol-

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ith size rather than (just) mass. For example, Fig. 2b shows the linear relationship between size and mass – it is obvious from this that what the study by Horigome et al. shows is a primary change in coccolith size across biogeographic regions.

Author Response: The data presented in figure 2b of our manuscript are an average of the raw data obtained for each of the 70 studied sites. Our data clearly reflect more a change of coccoliths length rather than their calcite mass. However, when we consider the raw data (see figure R1), it appears that the variability is higher for the mass than for the length. One could argue that this variability would be related to our methodology. Indeed, the coccoliths being measured through cross-polarized light, and their orientation with respect to the Crossed-Nicols may change the mass. However, Beaufort (2005) showed that the mass for a single *E. huxleyi* coccolith varies by less than 1 pg when the orientation changes from 0° to 90° (see figure 4 in Beaufort, 2005). This variation of 1 pg is far below the variations that we observed for coccoliths of the same size in our raw data. This suggests a higher variability in the mass of calcite than in the length.

3) Methodology. A recent paper by Bollmann, also in Biogeoscience Discussions (10, 11155-11179, see full reference below) now appears to question the underlying methodology used in the study by Horigome et al. – specifically the calibration used and the ease of identifying *E. huxleyi* coccoliths using this method. The authors will need to address the methodological issues raised by Bollmann (2013) to validate their own findings. (Bollmann J (2013) Technical note: Weight approximation of single coccoliths inferred from retardation estimates using a light microscope equipped with a circular polariser – (the CPR Method), Biogeoscience Discussions 10, 11155-11179).

Author Response: The new method described by Bollmann (2013) in BGD offers a new, interesting, and promising approach for calculating coccolith carbonate weight. Both methods (this study and that of Bollmann (2013)) are useful tools and have both strengths and limitations. As requested by Poulton and colleagues, we wish here to clarify our methodological approach and elaborate on the points raised by Bollmann.

Full Screen / Esc

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Discussion Paper



One of the main issues raised is that the calibration performed using our method relies on the use of inappropriate pure crystalline calcite particles. The author argues that particles of 1 to 5  $\mu\text{m}$  are outside the valid range of 0 – 1.55  $\mu\text{m}$  thickness proposed by Beaufort (2005). This statement would be true if the considered particles were spherical. But, the particles that were used by Beaufort (2005) and in this study have an elongated shape as shown in Figure R2, making those particles appropriate for the calibration (i.e. within the valid range of 0 – 1.55  $\mu\text{m}$ ).

Another point is related to the accuracy of the weight transfer function calibration. This is potentially limited by the random particle orientation on the slide and not showing necessarily their maximum interference colour/grey value as they are randomly distributed with respect to the Crossed Polariser. This is an important points that it is continuously tested and validated. It is important to note that coccolith crystals are built very regularly with respect to the orientation of their calcite elements and are aligned in a circle, leading to the extinction cross. It could be argued that a randomly orientated calcite powder would have the same proportion of the powder in extinction as the elements in a coccolith. When the powder is fine enough the slight differences due to different orientations could be minimized.

When comparing the data of *E. huxleyi*/small placoliths ( $n=70$ ) presented in figure 2A (Bollmann, 2013) to our raw data ( $n=10333$ ) (Figure C1), it is clear that the measurements performed by Bollmann (2013) do not cover a wide range of natural variability of coccoliths' morphometry. This is most likely due to the low number of specimen measured in his study. However, it is interesting to note that  $\sim 74.5\%$  of the specimens ( $\sim 7700$ ) measured in our study are in agreement with the values obtained by Bollmann (2013) if we take into account the accuracy of his method (see light grey boxes in the Figure C1).

Finally, the method developed by Beaufort (2005) and used in our study, in most cases, is in good agreement with others methods (Bollmann, 2013, Young and Ziveri, 2000) to estimate the calcite mass of an individual coccolith.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

4) What about morphotypes? It is well recognised in the literature that several morphotypes of *E. huxleyi* exist, and these appear to have clear biogeographical distributional patterns with variations in coccolith size and calcite content. There is also considerable intra-type variability in coccolith size (within one cultured strain) which has been examined in culture studies (e.g., Paasche 2002 *Phycologia*). The authors extensively list how morphometry (coccolith biometry) has been linked with environmental factors (pg 9297, ln 29 – 9298, ln 1 - 3). However, only two of these references include field data and there are several key missing references on morphotypes in this section (e.g., Cubellis et al. 2007 MEPS; Cook et al. 2011 *J Phycology*; Poulton et al. 2011 MEPS; Smith et al. 2012 PNAS; Hendericks et al. 2012 MEPS). pg 9298, ln 3-4: “the observed changes in *E. huxleyi* calcite mass distribution could be controlled by ecological preference of the different morphotypes”. Presently, the literature would support this observation, but the authors do not link the changes in coccolith size they detect with the distribution of morphotypes. If the authors have access to the SEM images from the sites (as shown in Fig. 1) why is this not examined in more detail or the study’s findings presented in this context? These patterns potentially explain the variations in coccolith size that they observe. Fig. 1 has images of different morphotypes from the different areas, but there is very little discussed in the paper recognizing these differences.

Author Response: It is correct that several *E. huxleyi* morphotypes have been described in recent years and for some of these the biogeographical distributions have been better defined for some areas (although in general, the distribution patterns of the different *E. huxleyi* morphotypes are still not completely known). However, all the mentioned studies (including Boeckel and Baumann, 2008) dealt with plankton samples and with coccosphere data. Those samples allow distinguishing between morphotypes, while it is more complicated in the sediments, where generally only single coccoliths and therefore much less identification parameters are available (since the coverage of the central area is often missing in *E. huxleyi* coccoliths of sediment samples). Therefore, only the subdivision into *E. huxleyi* > 4  $\mu\text{m}$  and *E. huxleyi* < 4  $\mu\text{m}$  have

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thus far often been used in sediment studies, whereas observations on morphotypes are still scarce (and, to our knowledge, have only been done by Boeckel and Baumann, 2008).

5) Have the oceanographic conditions over the last 12,000 yrs been stable in the study area? The authors provide no evidence or literature supporting the stability of temperature, salinity, nitrate and phosphate over the Holocene in the study area. Is it correct to assume that these have been stable in concentration and magnitude over this period? Have the frontal boundaries between the regions studied also stayed the same? As such, is it suitable to correlate annual values of these against an integrated Holocene record of coccolith size? Were the coccoliths deposited at the same rate throughout the Holocene in these areas? Also, why is chlorophyll-a included? Has chlorophyll-a stayed stable for the last 12,000 yrs?

Author Response: The sediments analyzed in this study cover a temporal interval ranging from ~1700 yr to ~150 yr (except for core MD02-2594, for which the core top is expected to be younger, cf. Martinez-Mendez et al., 2010). The South Atlantic has been recently the focus of several paleoceanographic studies mostly centered on the surface ocean temperature evolution, dynamics of the regional oceanic fronts, and upper ocean stratification during the glacial-interglacial and millennial-scale climate changes of the last deglaciation, last glacial period, and previous climatic cycles (e.g., Barker et al., 2009 – Nature; 2010 – Nature Geoscience; Scussolini & Peeters, 2013 – Paleooceanography), while relatively little (or no) attention has to date been paid to the ocean/climate variability that may have characterized the current Holocene interglacial. One high quality continental record from the Tristan da Cunha (Ljung et al., 2008 – Climate of the Past), however, tackles the South Atlantic climate variability during the Holocene and concludes that “a major change of the general circulation pattern took place around 8800 cal. yrs BP, likely reflecting the establishment of the present-day circulation regime, due to a meridional shift of the regional fronts”. This time span does include all of the samples analyzed in the present study (see above), implying that it is

Full Screen / Esc

Printer-friendly Version

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Discussion Paper

unlikely that major changes in the regional oceanic fronts and in the associated physicochemical properties may considerably bias our assumption of using modern surface ocean parameters to decipher the calcification controls on the coccoliths found in the surface sediments analyzed here.

6) Have the statistics been correctly interpreted? The cluster analysis of just coccolith mass (Supplementary Fig. S2) shows a very different pattern to that including both coccolith mass and environmental parameters (Fig. 3a). Fig. 3a shows a strong match to the distribution of water masses through the study area, indicating that the clusters are driven by hydrological factors, and that mass is a minor factor. No-where are the factors driving this clustering examined (e.g., Eigenvectors). The PCAs (Fig. 3c, 3d) show strongly how temperature (and salinity) is negatively correlated with nutrient concentrations (warm waters have low nutrients), but do not “reveal that more than 83% of the *E. huxleyi* calcite mass variance is explained by two factors” (pg 9296, In 21-22). The PCAs show that 83% of the variance between stations can be explained by two factors, which are an amalgamation of temperature-salinity-nutrients-chlorophylla-coccolith mass-carbonate ion and pH-pCO<sub>2</sub>. The relative degree to which coccolith mass (size) influences the PCAs is not shown. Furthermore, salinity and nitrate (and chlorophyll-a) appear as strong factors in the PCA, comparable with temperature, phosphate and pH-pCO<sub>2</sub>, but are surprisingly overlooked in the discussion. Why are these factors ignored? Where is the data to support the statement “carbonate ion concentration was also correlated with the first PCA factor, although according to our analysis it represents a minor contribution” (pg 9297, In 5-6)? How do the authors get to the conclusion (pg 9298, In 8-9) that “mass distribution is linked to surface water phosphorus and [the] temperature where coccolithophores calcify”? What happened to salinity, nitrate and chlorophyll-a?

Author Response: This is an excellent point and we do agree with the reviewers that the statistical analysis presented in our study can be improved. Hence, in the revised manuscript we will clarify/correct our analysis. First, we decided to slightly

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



change our approach as suggested also by J. Henderiks, (<http://www.biogeosciences-discuss.net/10/C5975/2013/bgd-10-C5975-2013.pdf>): as the aim of this study is to identify the environmental parameters that may influence the mass of *E. huxleyi* coccoliths retrieved in surface sediment samples, we first performed hierarchical cluster analyses (HCA) on the environmental parameters only (temperature, salinity, nitrate, phosphate, Chl-a, pH, pCO<sub>2</sub>, and [CO<sub>32</sub>-]) in order to highlight areas with different physicochemical properties. In a second step and for each area (cluster) resulting from the HCA, we performed principal component analysis (PCA) between the environmental parameters and the mass of *E. huxleyi* in order to reveal possible correlations.

We then present the different steps involved in the statistical analyses:

1- Data preparation - The final dataset of *E. huxleyi* mass was obtained by averaging the mass of all the specimens measured for each sample (an average of ~150 specimens were measured for each sample). - At each sample site, the environmental parameters (except Chl-a concentrations) were averaged from the surface to 50m water depth. All these parameters were extracted from different existing datasets presented in the manuscript. For the Chl-a concentrations, we used the annually averaged values obtained from satellite imagery, as described in the manuscript. - All the data were standardized before statistical analysis as follows: for a given sample and a considered parameter, the average of all the samples was subtracted and the result divided by the standard deviation of all the samples. The standardized values of *E. huxleyi* mass, temperature, salinity, nitrate, phosphate, chl-a, pH, pCO<sub>2</sub> and [CO<sub>32</sub>-] were used.

2- Hierarchical cluster analyses: - We used the Ward's Method (Ward, 1963) and squared Euclidian distance on the standardized values of the environmental parameters (temperature and salinity; nitrate, phosphate and chlorophyll a concentrations; and pH, pCO<sub>2</sub> and [CO<sub>32</sub>-]). We chose this method because it minimizes the total within-cluster variance (Ward, 1963). - In the revised manuscript, we finally decided to not use/present the results of the HCA on the mass of *E. huxleyi*. Indeed, in this case the clusters are distributed according to different classes of *E. huxleyi* mass ([1.73 pg;

Full Screen / Esc

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Discussion Paper



2.4 pg,] 2.4 pg; 2.95 pg]. . .). This classification does not allow any interpretation regarding the influence that the environmental parameters may have on *E. huxleyi* mass. - The results of the HCA on the environmental parameters show that the samples can be grouped into 3 main clusters: cluster #3 represents the subantarctic waters, cluster #2 represents the waters of the south Atlantic gyre and finally cluster #1 represents the waters flowing between these 2 structures (see revised figure S2). The highest gradients of temperature, salinity, nitrate, phosphate and [CO<sub>3</sub><sup>2-</sup>] are observed in cluster #3 (subantarctic area). The highest gradients of chlorophyll a, pH and pCO<sub>2</sub> are observed in cluster #1. Finally cluster #2 presents the lowest gradients of temperature, salinity, nitrate, phosphate and chlorophyll a.

3- Principal component analysis: As with the HCA, the PCA were conducted on the standardized values of the different parameters. The PCA included the environmental parameters and the mass of *E. huxleyi*. In a first step we performed PCA on the entire dataset to have a general view. Then PCA were conducted on the parameters of the 3 different clusters. - Results of the PCA conducted on the entire dataset show that ~80% of the variance within the samples can be explained by 2 factors. The first factor (F1), which explains ~60% of the variance, is driven (in order of importance) by the temperature (18% of F1), the phosphate (16.3% of F1), the nitrate (15.6% of F1), the salinity (15.3% of F1), the [CO<sub>3</sub><sup>2-</sup>] (14.7%), and the mass of *E. huxleyi* (2.7%). The second factor (F2) explains 20% of the variance within the samples and is driven by the pCO<sub>2</sub> (33.6% of F2), the pH (32.9% of F2) and the Chl-a (16.4% of F2), the others parameters having very low scores. An interesting result is that these analyses show that the mass of *E. huxleyi* presents a significant correlation (within a 95% confidence interval) with all the parameters, except the nitrate and the phosphate (see Figure S3 of the revised manuscript): the most important being the Chl-a ( $r=0.406$ ), the pCO<sub>2</sub> ( $r=-0.372$ ), the salinity ( $r=-0.359$ ), the pH ( $r=0.356$ ), the temperature ( $r=-0.305$ ) and the [CO<sub>3</sub><sup>2-</sup>] ( $r=-0.268$ ). This suggests a possible influence of the productivity, the carbonate system and the surface hydrography on the mass of *E. huxleyi*. However, further analyses are needed to better understand the role played by the environmental param-

eters on the mass of *E. huxleyi*. - Results of the PCA conducted on the samples of cluster #1 show that  $\sim 73.5\%$  of the variance within the samples can be explained by 2 factors. The first factor (F1), which explains  $\sim 43\%$  of the variance, is driven (in order of importance) by the temperature (22.9% of F1), the pH (20% of F1), the pCO<sub>2</sub> (19.5% of F1), the salinity (12.8% of F1) and the mass of *E. huxleyi* (8.5%). The second factor (F2) explains  $\sim 30.4\%$  of the variance within the samples and is driven by the nitrate (25% of F2), the phosphate (19.6% of F2), the Chl-a (18.8% of F2) and the [CO<sub>3</sub><sup>2-</sup>] (14.9% of F2). An interesting result is that these analyses show that the mass of *E. huxleyi* presents a significant correlation (within a 95% confidence interval) with the pH ( $r=0.621$ ), the pCO<sub>2</sub> ( $r=-0.605$ ) and the temperature ( $r=-0.537$ ) (see Figure S3 of the revised manuscript). This suggests a possible influence of the carbonate system and the temperature on the mass of *E. huxleyi*. - Results of the PCA conducted on the samples of cluster #2 show that  $\sim 75.6\%$  of the variance within the samples can be explained by 2 factors. The first factor (F1), which explains  $\sim 48\%$  of the variance, is driven (in order of importance) by the nitrate (17.2% of F1), the phosphate (16.9% of F1), the Chl-a (16.2% of F1), the mass of *E. huxleyi* (14.5% of F1) and the [CO<sub>3</sub><sup>2-</sup>] (10.5% of F1). The second factor (F2) explains  $\sim 27.7\%$  of the variance within the samples and is driven by the pCO<sub>2</sub> (21.8% of F2), the pH (21.7% of F2), the temperature (19.9% of F2) and the salinity (16.8% of F2). An interesting result is that these analyses show that the mass of *E. huxleyi* presents a significant correlation (within a 95% confidence interval) with the phosphate ( $r=-0.704$ ), the nitrate ( $r=-0.66$ ) and the Chl-a ( $r=-0.65$ ) (see Figure S3 of the revised manuscript). This suggests a possible influence of the nutrients on the mass of *E. huxleyi*. - Results of the PCA conducted on the samples of the cluster #3 show that  $\sim 90\%$  of the variance within the samples can be explained by 2 factors. The first factor (F1), which explains  $\sim 68.2\%$  of the variance, is driven (in order of importance) by the temperature (16% of F1), the nitrate (15.8% of F1), the phosphate (15.7% of F1), the salinity (15.5% of F1), the [CO<sub>3</sub><sup>2-</sup>] (15.3% of F1), the Chl-a (14% of F1) and the mass of *E. huxleyi* (7.4% of F1). The second factor (F2) explains  $\sim 22.5\%$  of the variance within the samples and is driven by the pCO<sub>2</sub>

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

(49.1% of F2) and the pH (48.4% of F2). An interesting result is that these analyses show that the mass of *E. huxleyi* presents a significant correlation (within a 95% confidence interval) with all the parameters except the pH and the pCO<sub>2</sub> (see Figure S3 of the revised manuscript): the most important being the nitrate ( $r=-0.632$ ), the phosphate ( $r=-0.62$ ), the temperature ( $r=0.609$ ), the salinity ( $r=0.562$ ), the Chl-a ( $r=0.557$ ) and the [CO<sub>32-</sub>] ( $r=0.554$ ). This suggests a possible influence of the surface hydrography, the nutrients and to a lesser degree the [CO<sub>32-</sub>] on the mass of *E. huxleyi*.

All these results are presented and discussed in the revised manuscript.

7) Where are the results? Relative to a 2-3 page introduction, a 4 page methods section (including a lengthy discussion of the birefringence method, which is published, and has been applied in numerous studies) and a 3.5 page discussion – why is the results part so short (1.5 pages). There is also no presentation of the raw (pre-statistical) results, apart from the difficult to interpret bubble plots in Fig. 1. Are these mean values?

Author Response: The results concerning the mass of *E. huxleyi* as well as the results obtained from the HCA and the PCA are now presented in detail in the revised manuscript. The raw data (those presented in the Figure R1) will be available as well. Figure 1 of the BGD manuscript was changed to avoid clutter.

8) What is the comparison with the Beaufort et al. (2011) paper all about? The authors state (pg 9297, ln 9) that the data compilation of Beaufort et al. (2011) was based on living coccolithophore water samples. However, only 24% (180 of 735 samples, not the number of coccoliths analysed) of the data in Beaufort et al. (2011) are water column samples, the other 75% are sediment trap material. Also, where did the authors get the Beaufort et al. (2011) data from? Some acknowledgement should be included?

Author Response: We discuss and compare the results obtained by Beaufort et al. (2011) since the main outcome of their study is that pCO<sub>2</sub> and [CO<sub>32-</sub>] should be seen as the main controlling factors on the *E. huxleyi* mass, while our results (obtained

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

employing the same approach) suggest that the surface hydrography and nutrient availability should be taken into account too. This is why we compare our results with their results in Fig. 4 of our manuscript. For the comparison we selected the water sample results presented by Beaufort et al. (2011) rather than the entire dataset, since the other samples were retrieved from sediment cores covering a longer period and a much larger carbonate chemistry range than our samples. Finally, no sediment trap material was used in the study of Beaufort et al. (2011), as suggested by Poulton and colleagues.

9) What about light? The authors do not include any information on relative light levels in the environments examined. This is despite several studies showing the importance of light on coccolithophore distribution (e.g., Charalampopoulou et al. 2011 MEPS), growth rates and cellular PIC:POC (Muller et al. 2008 L&O), and irradiance also modulates their response to pCO<sub>2</sub> (Zondervan et al. 2007 DSRIII). For a full examination of the environmental factors influencing coccolithophore growth and calcification, all factors should be included.

Author Response: In the present study, light penetration has not been considered in the evaluation of the environmental parameters relative to the *E. huxleyi* coccolith mass distribution. The data sets considered here are relative to surface sediments representing the photic zone export production and sedimentation, and not a distinctive water depth.

SPECIFIC COMMENTS - Abstract No quantitative results presented in the abstract, despite the strong conclusions. Where is the environmental data from?

Author Response: We do not see the abstract as the appropriate place for results, though it is more appropriate for over-arching conclusions.

- Introduction pg 9286, ln 26 – pg 9287, ln 1: Coccolithophores drive very little of the organic production in phytoplankton communities, and hence their photosynthesis is not the important term driving the PIC:POC ratio – it is their calcification.

Author Response: Coccolithophores are a minor contributor of total phytoplankton POC production. In the text mentioned in this comment, our intention was to highlight the fact that coccolithophores perform photosynthesis and produce large amount of marine CaCO<sub>3</sub>. The sentence is "They are responsible for the photosynthetic fixation of inorganic carbon, regulating the particulate inorganic-organic carbon ratio and a large portion of the calcium carbonate (CaCO<sub>3</sub>) production". We will modify this sentence such as : "They are fixing inorganic carbon through photosynthesis, regulating the particulate inorganic-organic carbon ratio mainly by producing a large amount of calcium carbonate (CaCO<sub>3</sub>)".

- pg 9288, ln 13-16: So diagenesis only occurs below the lysocline? The selection of sites situated above the modern lysocline may reduce the post-depositional effects of diagenesis, but certainly doesn't prevent them. Citing Boeckel et al. (2006) instead of Boeckel and Baumann (2008) would be more appropriate here. It should also be noted, however, that Boeckel et al. (2006) refer to Biscaye et al. (1976) for the position of the lysocline in the South Atlantic. Furthermore, Boeckel et al. (2006) clearly state that 'sediment assemblages preserved in samples from water depths of less than 4000 m might also be affected by dissolution due to change in alkalinity at the sediment-water interface' (section 3.3). SEM images are required to verify sample preservation.

Author Response: The preservation of the selected samples is generally good and has been documented by SEM work in Boeckel et al., 2006 and Boeckel and Baumann (2008). Assemblages preserved in samples from water depths less than 4000 m of course might be affected by dissolution, in areas of high TOC content, as at the continental margin of SW-Africa and thus in the periphery of the Namibian Upwelling. But in the present study most of the samples are from areas far away from the upwelling-influenced high-productive areas.

- pg 9291-9292, lns 28-1: *Gephyrocapsa oceanica* is not a "well studied common species" in the modern ocean. What is the argument for combining two species with coccoliths of very different masses?

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Author Response: *Emiliana huxleyi* and *Gephyrocapsa oceanica* are evolutionary related and within the most studied living species. There are several articles published on the physiology, ecology and paleoceanography of these species.

- Methods pg 9291, Equation 1 – where does “2275.14” come from? All the other terms are defined but not this one.

Author Response: The term “2275.14” refers to the slope of the linear regression presented in figure 2a of our BGD article. This will be added to the revised manuscript.

- pg 9292, In 24: Why include chlorophyll-a? This is surface chlorophyll-a, so lacks any depth resolution and misses potential deep chlorophyll maxima, which are important in the subtropics.

Author Response: The mean annual surface chlorophyll-a concentration derived from satellite is used in this study to compare relative quantification of primary production. The coccolith mass sedimentary record is a mediated values resulting from hundreds of years of export production and sedimentation.

- pg 9293, In 3-5: What control does chlorophyll-a have over coccolith ecology or calcification?

Author Response: The chlorophyll a does not have any control on the ecology of the coccolithophorids or their calcification. In this study, we used this parameter as an indicator of the surface productivity.

- pg 9293, In 13-14: How has *E. huxleyi* mass been included in the cluster analysis? As binned data, average values? Was it transformed/standardised?

Author Response: The mass values of all the specimens recognized for a given sample were averaged in order to have one value characterizing each sample. Then, for the cluster analysis as well as for the PCA, the values of the mass were standardized: for each sample the average of the mass of the entire dataset was subtracted and divided by the standard deviation calculated for the entire dataset. The same procedure was

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applied to all the other parameters (temperature, salinity. . .)

- Table 1: Have all of these been sampled? More than one of these samples has a depth greater than 4400 m, despite the statement on pg 9290, ln 13-14.

Author Response: The modern hydrographic lysocline is around 4300-4400 m in the South Atlantic (Broecker and Peng, 1982). Moreover, it has been shown that extremely high coccolith carbonate contents exist even down to 4700 m in the South Atlantic (Baumann et al., 2004; Baumann et al., 2004a)! Thus, coccoliths - or at least some species as *E. huxleyi* or the Gephyrocapsids - seem to be very resistant to dissolution (as already observed by Schneidermann, 1973 or by Berger, 1973)

- Results pg 9294, ln 23-24: How does lower coccolith calcite mass equate with lower abundances of *E. huxleyi*?

Author Response: Individual coccolith calcite mass are not necessarily related to coccolithophore abundance. However, we observed and documented this relationship for the clusters # 3 and 4. Since the statistics will be rerun in the revised version, we will remove the following text: "The spatial distribution of #3 and #4 might be due to the location of the Mid Atlantic Ridge, which can cause a considerable discrepancy of depths in a relatively short distance. In the central region of the Atlantic Ocean, especially at the Mid Atlantic Ridge, the samples present lower coccolith calcite mass, in line with previous studies that showed lower abundances of *E. huxleyi* (Boeckel et al., 2006)".

- pg 9297, ln 2: What is *E. huxleyi* production? Is it Primary production? Calcification? Calcite production? Production of cells (i.e., growth)? Coccolith production?

Author Response: In the sedimentary record, "*E. huxleyi* production" refers to the resulting product of *E. huxleyi* cell division and calcification rates and the coccospheres and coccoliths exported to the sea floor.

- pg 9297, ln 16: What is a 'fairly good correspondence' in the Boeckel and Baumann (2008) reference? Does this cover both subtropical and Antarctic comparisons?

**BGD**

10, C6632–C6659, 2013

Interactive  
Comment

Full Screen / Esc

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Interactive Discussion

Discussion Paper



Author Response: Boeckel and Baumann (2008) have "only" worked along a N–S transect from the South Atlantic Subtropical Gyre (8°S) to the Subantarctic Zone (45°S). So comparison is not possible for the southernmost samples, as well as for the samples around the southern tip of South Africa.

- pg 9297, In 10-11: Where are the statistics to prove that the data "clearly indicates very different trends"?

Author Response: The curves of the linear regressions characterizing the two datasets will be added to figure 4, showing that the relation between *E. huxleyi* mass and the [CO<sub>2</sub>] in our dataset is different from the one observed by Beaufort et al., (2011).

- Discussion pg 9298, In 24-26: What evidence is there that calcification rate determines thickness and growth rate determines size? It is not mentioned in Muller et al. (2008).

Author Response: The combined influence of different factors on calcification seems to be a more robust assumption than a sole parameter controlling marine calcification.

We will revise the sentence "In addition, the coccolith mass appears not to be a straightforward relation between cause and effect, in that it may reflect both calcification and growth rate, determining, respectively, the thickness and morphology (size) of the coccolith plates (cf. Müller et al., 2008)." Into: In addition, the coccolith mass appears not to be a straightforward relation between cause and effect, in that it may reflect both calcification and growth rate, determining the thickness and morphology (size) of the coccolith platelets."

Figures - The figures are generally difficult to interpret/see.

Author Response: All the figures have a high quality resolution. It is possible to zoom directly on them from a web browser or from the PDF.

- Fig. 1. Differences in bubble sizes almost impossible to see. Why include the SEM images when morphotypes are not classified or quantified? Technically the 'coc-

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

cololiths' mentioned are coccospheres.

Author Response: Fig. 1 is slightly changed in the revised manuscript in order to be more readable: a colored scale will replace the former bubbles. The pictures depict coccospheres and not coccoliths. The pictures are removed from the revised figure.

- How did the authors quantify a 'typical' coccolith?

Author Response: Here, the term typical rather refers to the fact that entire, well-preserved coccospheres were found in the sediment, rather than to a "typical" morphology of the coccospheres.

- Fig. 3. Very difficult to see colours and labels. What are the error bars in 3b? How do we interpret Fig. 3c? What are the important things to notice?

Author Response: Figure 3 will be split in the revised manuscript: the dendrogram (fig. 3a) will be placed in the supplemental material; only the map of the distribution of the clusters will be retained. Then for the figure 3b, all the environmental parameters will be compared to the mass of *E. huxleyi*. The error bars represent the 1 sigma error. Finally the results of the PCA will be presented in a new figure.

- Why is the cluster plot of coccolith mass (which the paper is all about) relegated to the Supplementary material?

Author Response: The paper is not only about the mass of *E. huxleyi*, but about the identification of the environmental parameters that govern the mass of *E. huxleyi* coccoliths. Then the HCA performed only on the mass of *E. huxleyi* is not enough to understand and explain the role played by the environmental parameters. This is why we focus our analyses and discussion on the results of the HCA performed on *E. huxleyi* and all the environmental parameters, and why the cluster plot of coccolith mass is relegated to the supplementary material.

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BGD

10, C6632–C6659, 2013

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C6652

**BGD**

10, C6632–C6659, 2013

Interactive  
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Discussion Paper



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#### FIGURE CAPTIONS:

Figure R1: Comparison between the measurements performed by Bollmann (2013) (red squares) and our study (open blue circles). The error bars accompanying Bollmann's series are extracted from his manuscript. The light grey boxes represent the full range of variability in agreement with his method. Please note that the regular spacing of our data along the X axis is only due to the resolution of our system which is 1 pixel ( $\sim 0.15\mu\text{m}$ ).

Figure R2: Image of 100 of the pure crystalline calcite particles that were used for the calibration. Please note that due to their shape the thickness of those particles are in

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the valid range of 0 – 1.57  $\mu\text{m}$  necessary for the method.

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Interactive comment on Biogeosciences Discuss., 10, 9285, 2013.

**BGD**

10, C6632–C6659, 2013

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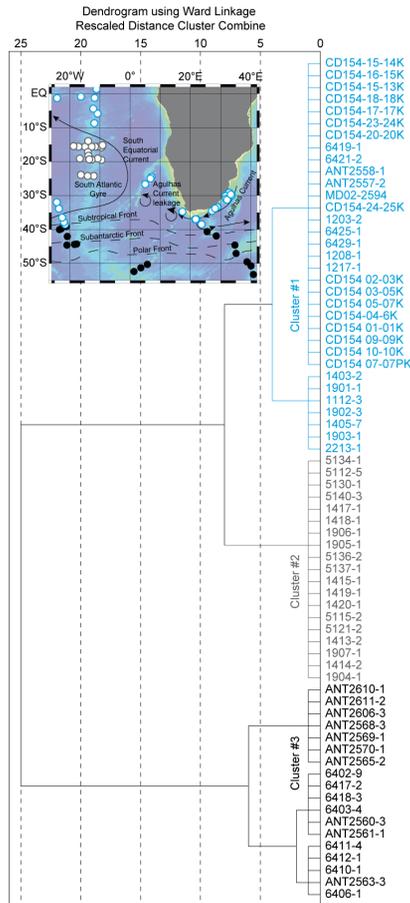
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Discussion Paper

C6655





# BGD

10, C6632–C6659, 2013

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Discussion Paper

Fig. 1. Figure S2 revised



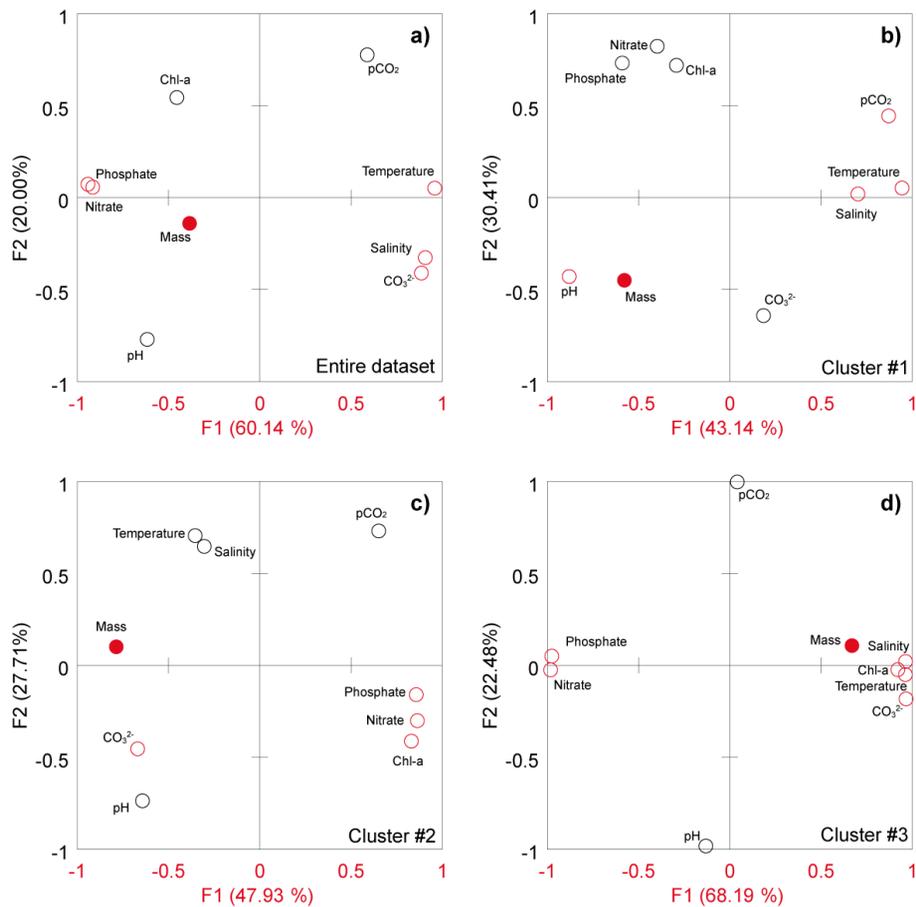


Fig. 2. Figure S3 revised

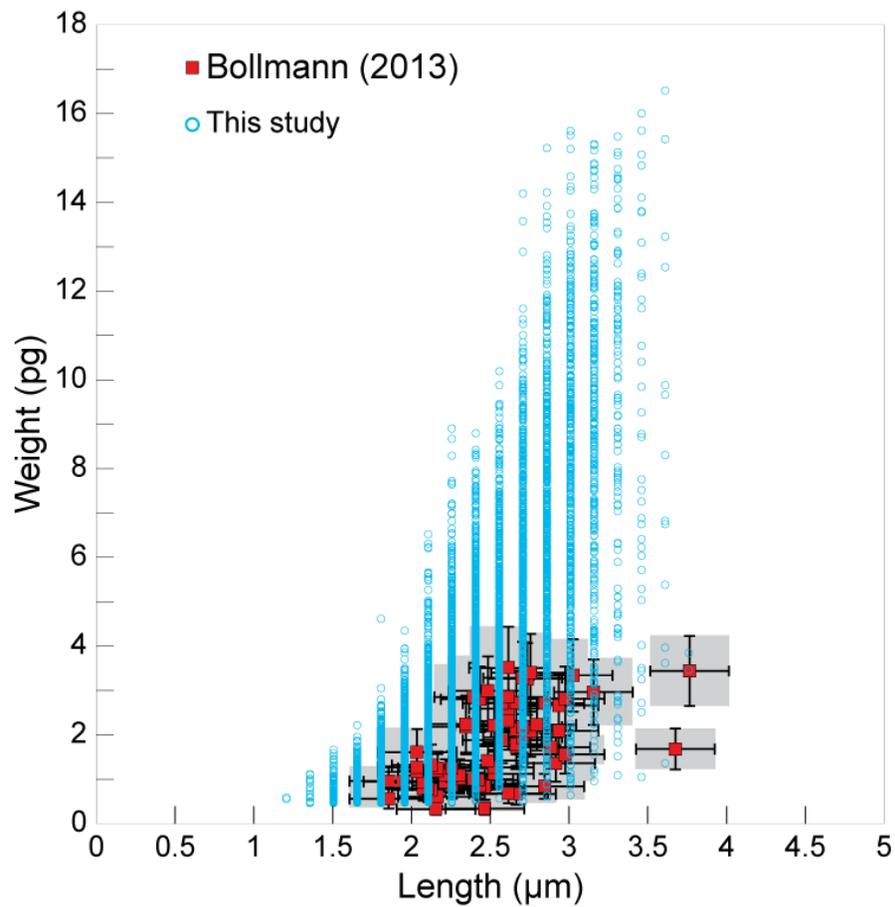


Fig. 3. Figure R1

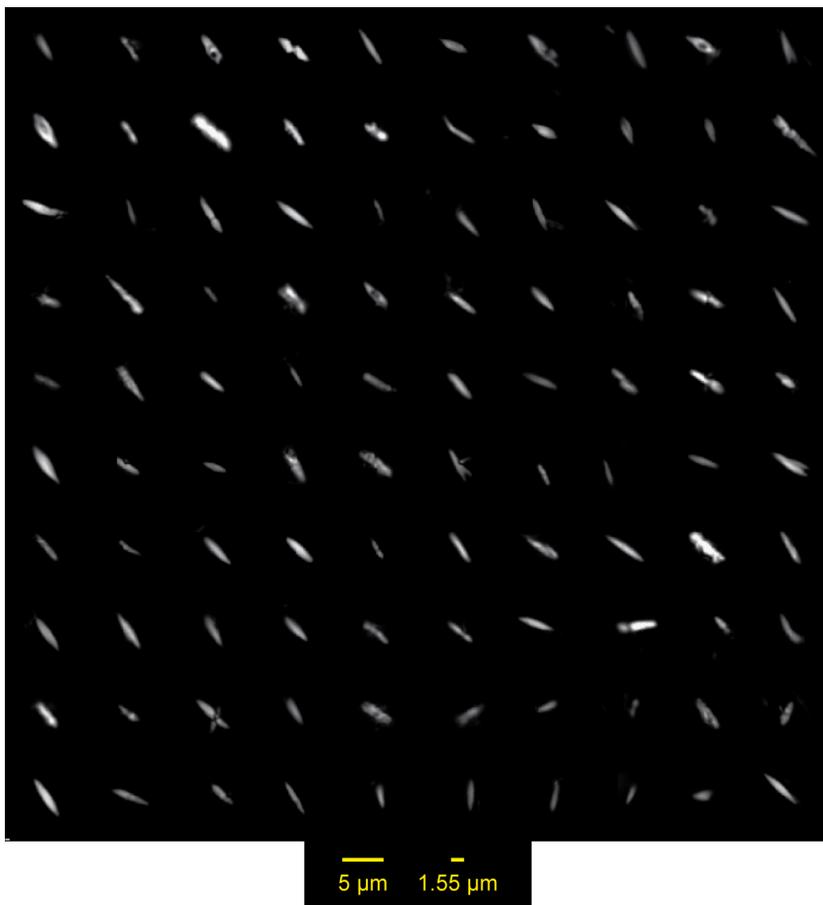


Fig. 4. Figure R2

C6659

## BGD

10, C6632–C6659, 2013

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