Original Reviews

From editor:

Comments to the Author:

Dear Author,
Thank you for submitting a reply to the referees' comments as well as a revised manuscript. There remains issues preventing acceptance of your manuscript for publication. I appreciate that you have added suitable caveats and recognized the limitations of your analysis and those in the original data. The fear that readers would blindly accept the pigment interpretation without reasonable understanding of the limitations has therefore been somewhat alleviated $\boxed{1}$ by the revised text. However, the list of 'supporting' references is misleading – many cite each other another. Support of the idea that the 19-hex pigment is a direct measure of coccolithophore biomass is not as strong as the manuscript implies. Another issue is the statistical significance as the increase relies on a limited part of the data set.

I am now in receipt of two referees reports (pasted below) which provide more context about the issues above. They also provide specific suggestions that I strongly encourage you to consider. Among them is the title which does not reflect the limitations of the study. This could be dealt with using one of the titles below but feel free to explore other options: $\boxed{2}$

- Apparent increase in coccolithophore abundance in the subtropical North Atlantic from 1990 to 2014

- Increasing coccolithophore abundance in the subtropical North Atlantic from 1990 to 2014?

Figures R1 to R3 are very useful to discuss the limitations. They should be provided as supplementary information so that you can refer to them in the text as needed. Finally, Biogeosciences strongly promotes the full availability of the data sets reported in the papers that it publishes in order to facilitate future data comparison and compilation as well as $\boxed{3}$ meta-analysis. This can be achieved by uploading the data sets in an existing database and providing the link(s) in the paper. Alternatively, the data sets can be published, for free, alongside the paper as supplementary information. The ascii (or text) format is preferred for data and any format can be handled for movies, animations etc?

Please provide a point-by-point reply to the comments provided in this second round of review. I hope that you will satisfactorily address all issues so that the review process could $\boxed{4}$ come to a timely end.

Regards,
Jean-Pierre Gattuso
Editor, Biogeosciences

Referee #1:

The revised paper now incorporates suitable caveats to make it clear the limitations of the inherent assumptions. However, I would like the authors to clear something up: there is a circular argument in some of the references they mention that should be removed or simplified, i.e. **5**

- Lomas and Bates (2004) – This reference does not implicitly prove that haptophytes or 19-hexanoyloxyfucoxanthin (19-hex) are directly related to coccolithophores, rather they reference Haidar and Thierstein (2001) to support this assumption. They state 'the Haptophyte group includes the coccolithophorids, for example, Emiliania huxleyi, that have been shown to be very important at BATS (H & R 2001)' and 'it is known that the Haptophytes present (at BATS) are dominated by Emiliania huxleyi that is on average > 5 μm (H & R, 2001).' Haidar and Thierstein (2001) never state that E. huxleyi, or the coccolithophores, dominate the haptophytes and make no direct comparison to pigments in this paper - they simply state that E. huxleyi dominates the coccolithophore community. **6**

- Steinberg et al. (2001) – This reference assumes all 19-hex comes from prymnesiophytes and that these are mostly coccolithophores. They provide no direct evidence to support this. Hence, using this reference to support the statement that 'coccolithophores comprise between 1% and 30% of chlorophyll a in subtropical gyre environments (this study-see Figure 2, Steinberg et al, 2001; Cortes et al., 2001)' assumes that all the 19-hex comes from coccolithophores. Please also note that Cortes et al. (2001) never looked at pigments in the context of their coccolithophore cell counts and also do not provide data to support this statement. (Also, the 'under bloom conditions they can approach 100% of the phytoplankton community' is not true – coccolithophore blooms incorporate many other phytoplankton groups, including diatoms and other flagellates (see e.g., Fernandez et al. 1993; Balch et al., 2014). **7**

- Dandonneau et al. (2006) – Show a very poor relationship (their Figure 14) of 19-hex and coccolithophore biovolume (in fact the abstract states this), with the relationship mostly being driven by their data from the Pacific North Equatorial Countercurrent (PNEC); their data in the South Pacific Subtropical Gyre shows no relationship when examined alone (see their Fig. 14). **8**

- Kavanaugh et al. (2014) – State 'We acknowledge the debate regarding the common usage of 19-hex to indicate oceanic coccolithophores because of its occurrence across multiple coccolith- and noncoccolith-forming taxa (Van Lenning et al. 2004). However, recent investigations have found good agreement (?) of 19-hex concentrations with coccolithophore biovolume (Dandonneau et al. 2006). Hence, this assumption is not independent of Dandonneau et al. (2006) and again shows no direct data that supports this assumption. **9**

- Riebesell et al. (2007) – This is based on mesocosms, where they do have pigment and coccolithophore cell counts, but never state in the paper that the two are equal: i.e., they talk about Prymnesiophyte pigments being predominantly from E. huxleyi. **10**

To conclude, the references cited by the authors include many inter-referencing and some incorrect information that should be clarified. None of the ‘numerous studies that agree with the conclusion that haptophytes are mostly coccolithophores’ provide direct data (apart from Dandonneau et al. 2006). The expanded explanation of how the authors have corrected for the different types of haptophyte pigments and the extended section on the limitations of the study are sufficient that readers of the paper do not blindly assume 19-hex is equivalent to coccolithophore biomass –referencing other studies which have done so does not help the argument (or the literature). **11**

Referee #2:

Most of my comments have been adequately addressed by the authors and several aspects like the uncertainties in deriving coccolithophore biomass from pigment data are discussed in more detail.

However, my main concern remains, i.e. there is only a statistically significant increase in Chla_hapto (Fig. 7a) from the early nineties onwards if the time series analysis is ending in 2012. No statistically significant increase is found for any other end year. Furthermore, pronounced (in terms of signal strength) and statistically significant trends, both negative and positive are found on shorter time scales, indicating some sort of cyclicity. This issue together with the uncertainties associated with deriving coccolithophore biomass from pigment data or satellite based particulate inorganic carbon estimates, should be clearly reflected both in the title (maybe consider adding a question mark) and the abstract, but also dicussed further in its overall implications. **12**

Another important aspect here is the observed magnitude of a potential trend. The authors should relate the calculated yearly concentration changes (please check the units in Fig 7a as they seem to be an order of magnitude too high, and also consider using the same units for Figs. 5,6 and 7) with average concentrations. Only then it becomes possible to judge whether an observed change, although potentially statistically significant is actually significant. **13**

The above comments on potential cyclicity versus long term trends and overall magnitude of such potential trend should also find consideration regarding the satellite products presented in figure 8. Furthermore, although the authors state that there is an overall statistically significant correlation of SeaWiFS and MODIS PIC data with Chla_hapto, the increase in latter during the last couple of years, which is actually responsible for the apparent long-term trend from the early nineties until 2012 (but also see comment above), is not at all reflected in the satellite data at BATS (compare Fig. 5). Thus, I do not see how the satellite is backing up the Chla_hapto data. **14**

ADDITIONAL COMMENTS:

1) The Schlueter et al. (2014) paper can not be used as a reference for coccolithophore calcification being insensitive to increasing CO2 in comparison to many other studies. First

of all, it is assessing adaptation potential as opposed to physiological acclimation, second, the 15
rates reported there are not directly comparable to other studies as not being the standard
amount of calcium carbonate per cell per day, and third, the restoration of calcification rates
(integrated over five days) to higher than present day levels was only found in a combined
CO2 and warming assay of the high CO2 high temperature acclimated cultures.

2) Concerning the potential DIC limitation of photosynthesis in coccolithophores in the
modern ocean, the authors mention a threshold of about 10 μmol/L. Fig. 2 in Riebesell et
al. (2004), from which they derived this value, highlights the problem I had with the notion
that the change in seawater CO2 during the last 20 years could directly be responsible for
changes in coccolithophore abundance by increasing growth rates. As shown in this Fig. 2 the 16
increase in photosynthetic rates when increasing seawater CO2 concentrations from about
10 to 15 μmol/L (equivalent to increasing atmospheric CO2 from a bout 250 to 400μatm
which is far greater than what would be expected for the last 20 years) is relatively small,
on the order of a few percent. And although there are other physiological studies showing
more pronounced decreases in photosynthesis below 10 μmol/L of CO2, the CO2 increase
during the last 20 years would be bound to minimal changes in photosynthesis.

3) The explanation for an opposing pattern in Synechococcus and Haptophyte abundances
described in the paper (Synechococcus is driving Haptophytes into DIC limitation) is not
convincing. If photosynthesis by other autotrophs would actually be able to provoke carbon
limitation in haptophytes why then is there only a opposing pattern with Synechococcus? 17
And secondly, it is again about the magnitude of a response. Both papers cited by the authors
(Riebesell et al. 2004 and Rost et al. 2003) show a very small response in photosynthesis
for reasonable (on the order of a few μmol/L) CO2 changes, expected for the open ocean.

Response to Reviewers and Proposed Revisions

We thank both reviewers and the editor for this second round of constructive comments. We hope we have adequately addressed all concerns with this revision.

Editor comments

$\boxed{\mathbf{1}}$ Thank you for your continued assistance with our manuscript. We have reduced the number of cited references (removing those that cite each other) and toned down the language in the Discussion section (Limitations of this study) regarding the published support for the assumption that 19-hex mainly comes from coccolithophores. We have also changed the title and abstract to reflect the variability in trends in Chl a from haptophytes (see specific responses below).

$\boxed{\mathbf{2}}$ We have changed the title to “Apparent increase in coccolithphore abundance in the subtropical North Atlantic from 1900 to 2014”. Thank you for the good suggestions.

$\boxed{\mathbf{3}}$ We have prepared a supplementary document containing these figures:

Figure S1: Mean Chl a hapto in the top 30 m versus coccolithophore cell counts from Haidar and Thierstein (2001).

Figure S2. Linear trends in mean NO_3^- concentration in the upper 30 m at BATS for a range of start and end years.

Figure S3: A version of Figure 5 with all unfiltered data points.

Figure S4: A version of Figure 6 with all unfiltered data points.

We cite these figures in the text. Figure S1 is cited in the Methods and Discussion section 4.3. Figure S2 is cited in the Discussion section 4.1. Figures S3 and S4 are cited in the figure captions for Figures 5 and 6, respectively.

Regarding the uploading of datasets, we appreciate the offer to upload the data. However, all data that we used in this study is already publicly available and therefore, we cannot re-distribute it. The link for the BATS data is in the text (first line of the Methods section) and the link for the satellite (PIC and Chlorophyll) data is in the Acknowledgements. According to our co-author, Dr. Nick Bates, the Director of Research at the Bermuda Institute of Ocean Sciences (where BATS data originates) links to the data should be sufficient. Also, researchers at Hydrostation S and BATS are in the process of implementing a new data management plan with the BCO-DMO (`http://www.bco-dmo.org/project/2124`) and OceanSITES (`http://www.oceansites.org/`), which will routinely pull data from the BATS FTP site.

$\boxed{\mathbf{4}}$ A point-by-point reply follows. Thanks again for your help with our manuscript.

$\boxed{5}$ We thank the reviewer for pointing out this issue with some of our references. We have corrected our citations to remove any circular referencing. We have also toned down the language regarding studies supporting the assumption that 19'-hex is mainly from coccolithophores, since many of these studies make the same assumptions without supporting data.

In Methods:

"The result is the haptophyte chlorophyll a fraction, which in this study, we assume to be mainly from coccolithophores. Particularly at BATS, the dominant haptophyte group has been reported to be coccolithophores by, e.g., Lomas and Bates (2004) and Steinberg et al. (2001), but these references offer no direct evidence for this assumption. We did, however, find a significant correlation ($p < 0.0000001$, $r^2 = 0.69$) between coccolithophore cell counts published in Haidar and Thierstein (2001) and calculated Chl a from haptophytes at BATS (Figure S1). More generally, Dandonneau et al. (2006) report that coccolithophores often dominate the haptophyte community in open ocean environments, such as BATS."

In Discussion:

"There are several caveats of this study that must be discussed before these results can be put into context. First, a primary assumption in this paper is that the haptophyte group is mainly composed of coccolithophores. Though high haptophyte diversity has been reported in open ocean regimes (Liu et al., 2009), this does not necessarily contradict the assumption that coccolithophores are the dominant type of haptophyte. While several studies describe coccolithophores to be the dominant haptophyte at BATS (see Methods; Lomas and Bates, 2004; Steinberg et al., 2001), this claim remains unsupported by specific data. We use the significant positive correlation between Chl a_{hapto} and cell counts from Haidar and Thierstein (2001) to back up our assumption that Chl a_{hapto} is mainly representative of coccolithophores (Figure S1). However, as with any study that derives phytoplankton community composition from signature pigments, inherent uncertainties are associated with changes in pigment content within a phytoplankton group over time."

$\boxed{6}$ We acknowledge that Lomas and Bates (2004) and Steinberg et al. (2001) do not prove the relation between 19'-hex and coccolithophores. However, given the familiarity of the authors with the BATS environment, we felt these references were worth citing.

$\boxed{7}$ We acknowledge that our claim (in the 'Responses to Reviewers' document) of coccolithophores comprising 1% – 30% of chlorophyll rests on the assumption that 19'-hex is mainly from coccolithophores. However, the other reference, Cortes et al. (2001) estimated that coccolithophores comprise 30% of the total community based on cell counts. Given the limitations of measurements (either cell counts or pigment analyses), it is difficult to assign exact percentages of phytoplankton contributions to primary production, which is why, in the Introduction, we simply say "coccolithophores generate a substantial fraction of primary production".

We thank the reviewer for providing references on contributions of coccolithophores to phytoplankton community composition under bloom conditions. We note that coccolithophore blooms consist of multiple species (not approaching 100% coccolithophores).

$\boxed{8}$ There is a poor correlation between 19'-hex and coccolithophore counts on a crude global scale, but the two open ocean sites (Pacific North Equatorial Current and South Pacific Subtropical Gyre) show a significant correlation ($r^2 = 0.6$). Both cell counts and 19'-hex concentration are low in the South Pacific Subtropical Gyre. Dandonneau et al. (2006) state "The coccolithophorids often dominate the Haptophytes in the open ocean." We cite this reference as evidence that coccolithophores are often the dominant haptophytes in open ocean environments. In the manuscript, we added the word "often" when referring to this study to emphasize that this is a generalization.

$\boxed{9}$ Yes, we acknowledge that using this reference is circular since this study cites the Dandonneau study. We have not included this reference in the manuscript.

$\boxed{10}$ This is a good point. Since Riebesell et al. (2007) used both cell counts and pigment data we assumed they had reasoning behind the assumption that the prymnesiophytes are mainly E. huxleyi, but they do not present specific evidence.

$\boxed{11}$ We appreciate that the reviewer accepts our revisions highlighting the limitations of using the pigment data as a proxy for coccolithophores. We modified the sections discussing these issues to clarify which studies are making this assumption without relying on specific evidence. We added references to Figure S1 (which shows mean Chl a hapto in the top 30 vs. coccolithophore cell counts from Haider and Thierstein, 2001) in these sections, which is the only hard evidence behind for our assumption.

$\boxed{12}$ We agree that the title and Abstract should reflect these limitations of our study.

To acknowledge variability in trends in Chl a hapto, we have changed the title to "Apparent increase in coccolithophore abundance in the subtropical North Atlantic from 1990 to 2014", as per the editor's suggestion. Also, we have added text to the Abstract to point to the variability:

"In this study, we use coccolithophore (Haptophyte) pigment data collected at the Bermuda Atlantic Time-series Study (BATS) site together with satellite estimates (1998–2014) of surface chlorophyll and particulate inorganic carbon (PIC) as a proxy for coccolithophore abundance to show that coccolithophore populations in the North Atlantic Subtropical Gyre have been increasing significantly over the past two decades. Over 1991–2012, we observe a 37% increase in euphotic zone-integrated coccolithophore pigment abundance at BATS, though we note that this is sensitive to the period being analyzed."

We report the sensitivity of these trends to different start and end years in section 3.4 of the manuscript. We have further added a sentence to section 4.2 to reflect the fact that there were periods of low Chl a hapto abundance:

"PIC, on the other hand, shows mainly positive trends over the whole gyre, in agreement with data from BATS – Chl a_{hapto} *shows overall increasing trends, despite some periods of low abundance. "*

$\boxed{\mathbf{13}}$ We thank the reviewer for this comment. Indeed there was a typo in the figure caption indicating when a star is put on the Chl a hapto plot (it was 0.2 μg m^{-3} when it should have been 2 μg m^{-3}). All trends without hatch marks are significant at the 95% level ($p < 0.05$). The stars were placed on certain trends to indicate when the trends were particularly strong, although they do not have a specific statistical meaning. Therefore, all boxes without hatch marks in Figure 7 indicate statistically significant trends.

We prefer to keep integrated Chl a hapto values in Figures 5 and 6 because we compare them to PIC and Chl a from satellite data, respectively. Since the satellite "sees" the water surface, the integrated pigment measurements better corresponded to the satellite measurements (correlations reported in section 3.5). We presented trends in mean Chl a hapto and Chl a concentrations in the top 30 m in Figure 7 so we could better compare trends between these pigments and the trends in average (top 30 m) concentration of, e.g., HCO_3^-.

$\boxed{\mathbf{14}}$ The PIC data presented in Figure 5 is just for one gridcell nearest to the BATS site. We discuss the lack of an increase in PIC in this gridcell in the "Limitations of this Study" section in the Discussion. The PIC satellite data backs up the Chl a hapto data due to the predominance of positive trends (Figure 8) for both SeaWiFS and MODIS, even if the individual gridcell closest to BATS does not show a strong increase at the end of the time series.

With respect to the potential cyclicity, we mention that there are some periods of low abundance in Chl a hapto when discussing the trends in section 4.2. We hesitate to describe a cyclic nature of high Chl a hapto abundance versus low abundance because the time series is not long enough to robustly support this observation. It will certainly to be interesting to see how Chl a hapto evolves in the near future at BATS, however!

$\boxed{\mathbf{15}}$ Point taken. We have changed the sentence to better reflect the results of this study.

"However, some recent studies show that coccolithophores increase calcification in response to increasing CO_2 *or restore calcification rates after an adaptation period under increasing* CO_2 *and warming (e.g., Iglesias-Rodriguez et al., 2008; Schluter et al., 2014)."*

$\boxed{\mathbf{16}}$ We are posing possible explanations for the positive Chl a hapto-HCO3 observation. Given the literature on carbon limitation in coccolithophores (for an overview see Paasche, 2001) we thought this was a possibility worth discussing. We have modified the text to acknowledge that coccolithophores may only increase their growth rate slightly given the increases in DIC.

"Overall, however, the increase in DIC experienced at BATS during the past two decades

could lead to slightly increased growth rates of coccolithophores (Riebesell, 2004, Rost et al., 2003), perhaps bolstering their competitive ability within the phytoplankton community."

If this slight increase in growth rate from increases in DIC (coccolithophores can use both CO2 and HCO3 for photosynthesis; Rost et al., 2003) were to boost coccolithophore growth to higher than that of the Synechococcus group, then coccolithophores will outcompete Synechococcus. Competition for a ecological niche between species can be highly nonlinear. We acknowledge later in the Discussion that other environmental controls could easily become more important in determining overall coccolithophore abundance.

$\boxed{\textbf{17}}$ The Synechococcus group and haptophytes are the dominant phytoplankton components in the upper euphotic zone at BATS. As mentioned in the manuscript, the Synechococcus group could also contain Trichodesmium and high-light Prochlorococcus. The strong negative correlation between Chl a hapto and Chl a syn indicates they are competing for a similar niche. Small changes in DIC, nutrients, or temperature (or other environmental influences) could all combine to influence which phytoplankton group (Synechococcus or haptophytes) dominate at any given time at BATS. We have added a sentence to reflect this:

"Small changes in DIC, nutrients, pH, or temperature could, in combination, influence which phytoplankton group (Synechococcus or haptophytes) dominate at any given time at BATS."

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~~Increasing~~ Apparent increase in coccolithophore abundance in the subtropical North Atlantic from 1990 to 2014

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Abstract

As environmental conditions evolve with rapidly increasing atmospheric CO_2, biological communities will change as species reorient their distributions, adapt, or alter their abundance. In the surface ocean, dissolved inorganic carbon (DIC) has been increasing over the past several decades as anthropogenic CO_2 dissolves into seawater, causing acidification (decreases in pH and carbonate ion concentration). Calcifying phytoplankton, such as coccolithophores, are thought to be especially vulnerable to ocean acidification. How coccolithophores will respond to increasing carbon input has been a subject of much speculation and inspired numerous laboratory and mesocosm experiments, but how they are currently responding in situ is less well documented. In this study, we use coccolithophore (Haptophyte) pigment data collected at the Bermuda Atlantic Time-series Study (BATS) site together with satellite estimates (1998–2014) of surface chlorophyll and particulate inorganic carbon (PIC) as a proxy for coccolithophore abundance to show that coccolithophore populations in the North Atlantic Subtropical Gyre have been increasing significantly over the past two decades. Over 1991–2012, we observe a 37 % increase in euphotic zone-integrated coccolithophore pigment abundance at BATS, though we note that this is sensitive to the period being analyzed. We further demonstrate that variability in coccolithophore chlorophyll *a* here is positively correlated with variability in nitrate and DIC (and especially the bicarbonate ion) in the upper 30 m of the water column. Previous studies have suggested that coccolithophore photosynthesis may benefit from increasing CO_2, but calcification may eventually be hindered by low pH_T (< 7.7). Given that DIC has been increasing at BATS by ~ 1.4 µmol kg^{-1} yr^{-1} over 1991 to 2012, we speculate that coccolithophore photosynthesis and perhaps calcification may have increased in response to anthropogenic CO_2 input.

1 Introduction

Coccolithophores are the most abundant type of calcifying phytoplankton in the ocean. Belonging to the phytoplankton group known as haptophytes, coccolithophores generate a substantial fraction of the primary production in many diverse marine environments from cold, sub-polar waters to warm, tropical waters (Thierstein and Young, 2004). Coccolithophores produce calcium carbonate ($CaCO_3$) shells that sink to the deep ocean forming chalk deposits and thus are important for global biogeochemical cycling of carbon and climate feedbacks. Further, coccolithophores comprise the base of many marine food webs and are widespread throughout the world ocean (Boyd et al., 2015). Changes in coccolithophore abundance could therefore have far-reaching effects from the ecosystem level to global carbon cycling (Boyd et al., 2015).

Ocean acidification from the gradual oceanic absorption of anthropogenic CO_2 has been projected to impact future coccolithophore populations, possibly disrupting the formation and/or dissolution of their $CaCO_3$ shells (e.g., Doney et al., 2009; Mackey et al., 2015). As CO_2 is absorbed from the atmosphere, it reacts with water releasing hydrogen ions (H^+) and increasing dissolved inorganic carbon (DIC). Excess H^+ ions and more DIC in the water column lead to a decrease in the carbonate ion (CO_3^{2-}) concentration in the ocean (for an overview of CO_2-carbonate chemistry see Bates et al., 2014). Lower CO_3^{2-} concentrations decrease the saturation state of $CaCO_3$ and could result in lower calcification rates in coccolithophores. Recently, Bach (2015) proposed that a substrate-inhibitor ratio may be a better indicator of potential biocalcification rates. Bicarbonate ions (HCO_3^-) are the substrate for calcification in most calcifying organisms, but high concentrations of H^+ ions can limit calcification. Therefore, a decreasing $[HCO_3^-]/[H^+]$ ratio may eventually hinder calcification, rather than low CO_3^{2-} concentrations. Even so, other factors (e.g., nutrient and light limitation) could exert a stronger control on calcification than carbonate chemistry (Bach et al., 2015). A CO_3^{2-} saturation state of less than one would still, however, cause the dissolution of $CaCO_3$ shells. Speculation of how coccolithophores will respond to increasing DIC and acidification (i.e., the balance between $CaCO_3$ production vs. dissolution) has been the subject of many laboratory and mesocosm studies (e.g., Iglesias-Rodriguez et al., 2008; Schlüter et al., 2014; Riebesell et al., 2007). These, however, have yielded mixed results, highlighting the complexity of biological responses to these changing oceanic conditions.

Numerous laboratory studies indicate that acidification of oceanic waters leads to a decrease in calcification rates for coccolithophores (Riebesell et al., 2000; Sciandra et al., 2003; Delille et al., 2005). However, some recent studies show that coccolithophores ~~have no response or even~~ increase calcification in response to increasing CO_2 or restore calcification rates after an adaptation period under increasing CO_2 and warming (e.g., Iglesias-Rodriguez et al., 2008; Schlüter et al., 2014). Indeed, responses to elevated CO_2 by different species of coccolithophores vary in all directions, making the extrapolation of these laboratory results to natural populations challenging. How calcifying phytoplankton will react to continually increasing CO_2 may vary from region to region in the world's oceans, depending on phytoplankton assemblages (e.g., the dominant species or strain of coccolithophore), available nutrients, and temperature (Sett et al., 2014). Coccolithophore responses to ocean acidification may be species specific or even vary within species (different *morphotype* responses, see Beaufort et al., 2011).

Differences in physiological mechanisms could play an important role in determining relative phytoplankton abundances under increasing DIC and acidification. For instance, nutrient uptake rates vary between phytoplankton species depending on the affinity of the transport mechanism for its substrate (e.g., phosphate ion). Laboratory experiments on the widespread coccolithophore *Emiliania huxleyi* have shown this species to have an efficient phosphate uptake system with a low half-saturation constant for phosphate, making it a superior competitor in phosphate-limited oceanic regions (Riegman et al., 2000). These results have been supported by field data from the subtropical Pacific Ocean (Cortés et al., 2001) and through a combination of field data and modeling in the NE Atlantic Ocean (Tyrrell and Taylor, 1996). For inorganic carbon uptake for photosynthesis, however, *E. huxleyi* displays a relatively high half-saturation constant compared to other phytoplankton (Riebesell, 2004; Rost et al., 2003), indicating that coccolithophores may benefit from increasing atmospheric CO_2 absorbed into the ocean. However, there are few in situ or observational studies of coccolithophore responses to increasing anthropogenic carbon (e.g., Feng et al., 2009; Freeman and Lovenduski, 2015).

To test the hypothesis that coccolithophores may be responding positively to additional CO_2 inputs, we employed data from the Bermuda Atlantic Time-Series (BATS), a long running oceanic time series in the North Atlantic Subtropical Gyre (Sargasso Sea) located at approximately 31.7° N, 64.2° W (Fig. 1). At this site, twice-monthly and monthly hydrographic and biogeochemical measurements have been made since the late 1980s (Fig. 1; Lomas et al., 2010; Bates et al., 2014). The BATS environment is characterized by Ekman down-welling and convergence, which results in an oligotrophic setting (Sarmiento and Gruber, 2006). While this area displays strong summer stratification (mixed layer depth $\sim 35\,\mathrm{m}$), seasonal overturning results in a deep mixing of the water column during winter (mixed layer depth $\sim 250\,\mathrm{m}$; Steinberg et al., 2001; Lomas et al., 2013). Though oligotrophic oceanic gyres have relatively low productivity compared to other areas of the oceans, they cover vast areas and thus are important on a global scale (Sarmiento and Gruber, 2006). Furthermore, the strong summer stratification experienced in these regions could be indicative of future trends, as increased stratification of the water column is projected with global warming (Gruber, 2011; Giovannoni and Vergin, 2012; Cabré et al., 2015). Thus, understanding phytoplankton dynamics subject to these environmental conditions is essential for accurately forecasting future ocean biogeochemistry.

Pigment analyses have been used to study the distribution, relative abundance, and assemblages of natural phytoplankton populations. Using high performance liquid chromatography (HPLC) to identify the presence and concentration of signature pigments, researchers can obtain relative components of chlorophyll *a* (Chl *a*) from phytoplankton (Letelier et al., 1993; Wright and van den Enden, 2000; Van Lenning et al., 2004). Coccolithophores, a haptophyte algae, are identified using signature pigments for haptophytes (mainly 19'-hexanoyloxyfucoxanthin), of which coccolithophores are likely the main component (Dandonneau et al., 2006), particularly at BATS (see Methods, Haidar and Thierstein, 2001; Steinberg et al., 2001; Lomas and Bates, 2004). While HPLC pigment analyses can provide a site-specific record of phytoplankton relative abundance, satellite-based records can provide information at larger spatial scales.

Ocean color remote sensing, through the Sea-viewing Wide Field-of-view Sensor (SeaWiFS; 1997–2010) and the Moderate Resolution Imaging Spectroradiometer (MODIS) Aqua (2002-present) platforms, has revolutionized our understanding of the ecological processes of the upper ocean on a variety of spatial and temporal scales. Satellite-estimated Chl *a* concentration has been used as a proxy for phytoplankton abundance and biomass since 1978. While most phytoplankton are not very effective light scatterers relative to their surroundings, coccolithophores produce $CaCO_3$ shells that are highly reflective. An understanding of coccolithophore-specific water-leaving radiances and the calcite-specific backscattering cross section allows for the concentration of coccolithophore particulate inorganic carbon (PIC) to be estimated via remote sensing (Gordon et al., 2001; Balch et al., 2005; Balch and Utgoff, 2008). Therefore, satellite estimates of PIC provide a qualitative proxy for coccolithophore abundance.

In this study, we combine pigment data from BATS along with PIC and Chl *a* measurements from the satellite record to assess recent trends in phytoplankton dynamics in the North Atlantic subtropical gyre, with a focus on coccolithophores. This data suggests that coccolithophore populations in the North Atlantic are increasing. Correlations indicate that they may be responding positively to increasing inorganic carbon from anthropogenic inputs in the upper mixed layer.

2 Methods

2.1 Data sources

Pigment measurements were obtained from the BATS website (http://bats.bios.edu), resampled at regular monthly intervals using a linear interpolation between measurements, and converted to relative Chl *a* components from different phytoplankton groups as in Letelier et al. (1993). Briefly, each phytoplankton group is associated with signature pigments that have relatively constant ratios with total Chl *a*. Signature pigment concentrations from each phytoplankton group, obtained via HPLC analysis, were converted to Chl *a* concentration using these ratios. This method has been verified in the North Pacific (Letelier et al., 1993) and the North Atlantic subtropical gyres (Andersen et al., 1996). We focused on measurements from the upper water column (top 30 m), consistently within the mixed layer (Steinberg et al., 2001; Lomas et al., 2013), but also examined trends and variability integrated over the depth of the euphotic zone (∼140 m) to verify congruence with the top 30 m of the water column.

Haptophyte pigments specifically were calculated using 19'-hexanoyloxyfucoxanthin (19'-hex) pigments and 19'-butanoyloxyfucoxanthin (19'-but). Coccolithophores and other prymnesiophytes contain 19'-hex and negligible amounts of 19'-but, while some other phytoplankton (e.g., chrysophytes and *Phaeocystis*) contain significant amounts of both 19'-hex and 19'-but (Zapata et al., 2004; Letelier et al., 1993). Based on the relative concentrations of 19'-but to 19'-hex measured at BATS, we subtracted out the phytoplankton pigment contribution that contains both 19'-hex and 19'-but, as in Letelier et al. (1993), and multiply the remaining 19'-hex concentration by a 19'-hex to Chl *a* ratio found in calcifying haptophytes (Letelier et al., 1993). The result is the haptophyte chlorophyll *a* fraction, which in this study, we assume to be mainly from coccolithophores. ~~This assumption is supported by several studies.~~ Particularly at BATS, the dominant haptophyte group has been reported to be coccolithophores by, e.g., Lomas and Bates (2004) and Steinberg et al. (2001)~~. Further, there is~~, but these references offer no direct evidence for this assumption. We did, however, find a significant correlation ($p < 0.0000001$, $r^2 = 0.69$) between coccolithophore cell counts published in Haidar and Thierstein (2001) and calculated Chl *a* from haptophytes at BATS (~~data not shown~~Figure S1). More generally, Dandonneau et al. (2006) report that coccolithophores often dominate the haptophyte community in open ocean environments, such as BATS.

In order to explore whether phytoplankton population dynamics are driven by carbonate chemistry parameters, we used the Mocsy Fortran 90 package (Orr and Epitalon, 2015) to solve the full carbonate chemistry system using available measurements at BATS (Bates et al., 2012). Using dissociation constants from Lueker et al. (2000), carbonate chemistry output from Mocsy agrees with other current carbonate system packages available (Orr et al., 2014). Input includes average concentrations of total alkalinity and DIC along with mean temperature and salinity in the top 30 m. Output includes pH_T, CO_3^{2-} concentration, bicarbonate (HCO_3^-) concentration, and aqueous CO_2 + carbonic acid ($CO_{2(aq)} + H_2CO_3 = H_2CO_3$*).

We used satellite observations of level 3, monthly binned PIC and Chl *a* from the Sea-viewing Wide Field-of-view Sensor (SeaWiFS; 1997–2007; limited data availability after 2007) and MODIS Aqua (2003–2014) on a 9 km (5 min) grid obtained from the NASA Ocean Colour distributed archive (http://oceancolor.gsfc.nasa.gov/). We calculated the mean satellite-derived PIC concentration in the BATS region that contains $> 95\,\%$ of pigment measurements (Fig. 1a).

2.2 Statistical analyses

2.2.1 Correlations

In order to identify correlations of phytoplankton pigment abundance across different species, with local environmental variables, and with satellite products, we performed correlation analysis on linearly detrended and deseasonalized (1 year boxcar smoothing) anomalies. Correlations between the main Chl *a* phytoplankton fractions present at BATS (*Prochlorococcus*, *Synechococcus*, haptophytes, and diatoms) were calculated for a variety of oceanic measurements hypothesized to influence phytoplankton abundance: DIC, alkalinity, inorganic nitrogen concentrations, temperature, salinity, and carbonate chemistry variables (see above section on carbonate chemistry). We also explored possible correlations between Chl *a* phytoplankton fractions and the mixed layer depth (MLD). We used two methods for calculating MLD. MLD_{sigma} was determined to be where the potential density anomalies (sigma-theta) at depth displayed a $> 0.125\,kg\,m^{-3}$ difference from surface waters, while MLD_{temp} was calculated as the location of 0.5 °C change in temperature from the surface (Monterey and Levitus, 1997). Mean density and temperature measurements within the top 10 m of the water column were used for "surface" values. We also tested correlations with the monthly mean North Atlantic Oscillation (NAO) index, obtained from NOAA National Weather Service Climate Prediction Center (http://www.cpc.ncep.noaa.gov/data/teledoc/nao.shtml).

2.2.2 Trends

In order to quantify temporal trends in pigment concentrations, biogeochemical measurements, and satellite data, we calculated the slope of a straight line that best fit the time-series in a least-squares sense. When comparing trends in pigments with trends in biogeochemical measurements at BATS, we used the average values over the top 30 m of the water column. Satellite Chl *a* and PIC trend analysis was performed on a grid cell basis.

3 Results

3.1 Chl a_{hapto} at BATS

Chl *a* from haptophytes (Chl a_{hapto}; assumed to be primarily from cocccolithophores, see Methods) is present throughout the euphotic zone at BATS and displays a pronounced seasonal cycle. Concentrations of Chl a_{hapto} surpassed 100 µg m^{-3} during periods of high abundance, with ~ 35 µg Chl a_{hapto} m^{-3} during during periods of relatively low abundance, such as between 2000 and 2004 (Fig. 1b). The bulk of haptophyte pigments occurred around ~ 80 m of depth, but pigments were also abundant in the upper 30 m, especially during spring. Haptophyte pigment concentration is low below depths of ~ 140 m. During the mid-90s and last six years of the dataset, Chl a_{hapto} was more concentrated, especially in the upper 30 m of the water column (Fig. 1b).

Haptophytes comprise roughly 30 % of the Chl *a* in the upper 30 m of the water column at BATS (Fig. 2), a percentage that persists from the start of measurements ($\sim$ 1990) to the end of our dataset ($\sim$ 2012). However, a period of low haptophyte abundance occurred between 2000 and 2004, reducing their relative contribution to Chl *a* to 15 %. *Synechococcus*

Chl *a* is variable, ranging from 20 to 70 % of the total Chl *a* in the upper 30 m. Unfortunately, signature pigment concentrations necessary to calculate *Synechococcus* Chl *a* were missing from the BATS dataset at the beginning of our time-series as well as during a five-year segment from ∼ 1997 to ∼ 2002 (shown as hatched area in Fig. 2). In contrast to generally high *Synechococcus* and haptophyte pigment abundance, *Prochlorococcus* and diatom pigments contribute relatively small fractions to the total Chl *a* in the upper 30 m. Correlations with potentially influential oceanographic drivers can aid to explain variable abundance of different phytoplankton Chl *a* fractions in these surface waters.

3.2 Correlations of Chl *a* components with oceanographic measurements at BATS

Correlation coefficients between detrended, deseasonalized anomalies of Chl a_{hapto}, Chl *a* from *Synechococcus* (Chl a_{syn}), Chl *a* from *Prochlorococcus* (Chl a_{pro}), and Chl *a* from diatoms (Chl a_{diatoms}) and anomalies in other oceanographic measurements/indices are presented in Fig. 3. Chl a_{hapto} shows strong positive correlations with DIC and HCO_3^- (explaining nearly 20 % of the variability). Chl a_{pro} showed similar, but somewhat weaker, correlations with DIC and HCO_3^-. Conversely, Chl a_{syn} displayed a strong negative correlation with DIC and HCO_3^- (Fig. 3). All phytoplankton pigment groups, except *Synechococcus*, were positively correlated with nitrate (NO_3^-) variability (measurements also include nitrite, NO_2^-; $NO_3^- + NO_2^-$ is referred to hereinafter as NO_3^-). CO_3^{2-} concentration, the saturation state of aragonite (Ω_{arag}), and temperature were negatively correlated with Chl a_{hapto}, opposite to Chl a_{syn}. Indeed, *Synechococcus* and haptophyte pigments display inverse correlations for nearly every variable tested, including temperature (Fig. 3).

Temperature is negatively correlated with Chl a_{hapto} (as well as Chl a_{pro} and Chl a_{diatoms}) and positively correlated with Chl a_{syn} (Fig. 3). Further, when the North Atlantic Oscillation (NAO) index is in a positive phase, temperatures over this region of the North Atlantic are generally warmer (Visbeck et al., 2001). Therefore, in line with the temperature correlations, the NAO index is negatively correlated with Chl a_{hapto}, opposite again to Chl a_{syn}. Mixed layer depth (MLD), an indicator of both temperature and nutrient availability, shows corresponding correlations with Chl *a* components. When the MLD is deeper, there is more

Chl a_{hapto} and less Chl a_{syn}. Both methods of calculating MLD (MLD_{temp} and $\text{MLD}_{\text{sigma}}$) showed similar correlations (Fig. 3).

3.3 Fluctuations in chlorophyll *a* from different phytoplankton groups

Haptophyte and *Synechococcus* pigments generally show opposing correlations with the variables tested (Fig. 3). This is supported by the opposing dominance of either *Synechococcus* or haptophyte Chl *a* throughout the timeseries (Fig. 4). Indeed Chl a_{hapto} and Chl a_{syn} show a significant negative correlation ($p < 0.00001$). During periods of low haptophyte pigment bundance (e.g., 2000–2004), *Synechococcus* pigments dominate the water column. Later in the time-series, however, *Synechococcus* pigments decline and haptophyte pigments increase. *Prochlorococcus* pigments (mostly low-light *Prochlorococcus*, see Discussion) at BATS reside mostly in the deep chlorophyll maximum at ~ 100 m of depth and are a relatively minor component of the Chl *a* in the upper 30 m of the euphotic zone. Diatom presence, usually associated with cold, high nutrient environments, is sporadic and generally low in this oligotrophic oceanic region of the subtropical North Atlantic, according to their signature pigments (Fig. 4).

In general, pigment analyses indicate that *Synechococcus* and haptophytes are the most abundant phytoplankton groups at BATS (Figs. 2 and 4). However, the category classified as "other" (includes chrysophytes, dinoflagellates, prasinophytes, among others) in Fig. 3 comprises a large portion ($> 30\,\%$) of the phytoplankton biomass during certain periods, indicating certain conditions may favor neither *Synechoccocus* nor haptophytes. The opposing correlations *Synechoccocus* and haptophyte pigments exhibit with DIC, HCO_3^-, CO_3^{2-}, NO_3^- and temperature could lead to interesting trends in phytoplankton abundance in an ocean increasingly influenced by anthropogenic climate change.

3.4 Trends in Chl a_{hapto} and total Chl *a* at BATS

A time-series of Chl a_{hapto} at BATS shows that haptophytes (mainly coccolithophores, see Methods), have been increasing significantly since 1990 ($p < 0.01$ for 30 m integral; $p <$

0.001 for 140 m integral; Fig. 5a, b). Mean concentration of Chl a_{hapto} in the upper 30 m of the water column has increased by 0.848 $\mu g\,m^{-3}\,yr^{-1}$ (standard error = 0.332), corresponding to a 68 % increase over the course of the BATS time-series (1991–2012; an overall increase of 17.8 $\mu g\,m^{-3}$), while the 140 m integral of Chl a_{hapto} has increased by 0.103 $mg\,m^{-2}\,yr^{-1}$ (standard error = 0.0307) corresponding to a 37 % increase (an overall increase of 2.2 $mg\,m^{-2}$). We assess the sensitivity of these trends to interannual variability by performing trend calculations for a range of start and end years over the time-series. The resulting trend pattern shown in Fig. 7a shows mostly positive trends in Chl a_{hapto}, except for end years in the 2000–2004 period. Total chlorophyll *a* (Chl a_{total}) also shows a significant positive trend over the time-series ($p < 0.05$ for 30 m integral; $p < 0.001$ for 140 m integral; Fig. 6; upper right corner of Fig. 7c). Figure 7c shows trends in Chl a_{total} for a range of start and end years, displaying a different pattern than that of the trends in the Chl a_{hapto} component of Chl *a*. For instance, for end years in the 2000–2004 period, Chl a_{total} shows positive trends (but nonsignificant), whereas Chl a_{hapto} shows significant negative trends (Fig. 7). Mean Chl *a* in the upper 30 m primarily exhibits negative trends in the later part of the time-series and most trends in Chl *a* are nonsignificant (Fig. 7c). Unfortunately, missing *Synechococcus* pigment data did not allow for long-term trend analysis of this group of phytoplankton (see hatched area in Fig. 2 and white area in Fig. 4).

In line with the results of our correlation analysis, the trends in HCO_3^- for various start and end years show a similar pattern to the trends in Chl a_{hapto} (Fig. 7b): mostly positive trends with slightly negative (but nonsignificant) trends in HCO_3^- concentration for end year in the 2000–2004 period, a low point in haptophyte pigments. Conversely, the trend pattern for the substrate-inhibitor ratio, $[HCO_3^-]/[H^+]$, is distinctly different from that of Chl a_{hapto}, exhibiting all negative trends (Fig. 7d). Trends in Chl a_{hapto}, assumed here to be mainly representative of coccolithophores (see Methods), can be further corroborated with particulate inorganic carbon (PIC) measurements from the satellite record.

3.5 Relating BATS pigment data to satellite products

Significant correlations were detected between Chl *a*$_{\text{hapto}}$ (30 m integral) measured at BATS and PIC derived from each satellite. The SeaWiFS-derived PIC correlated somewhat better with Chl *a*$_{\text{hapto}}$ than MODIS-derived PIC (SeaWiFS PIC-Chl *a*$_{\text{hapto}}$ $p = 0.0075$, $r^2 = 0.19$ vs. MODIS PIC-Chl *a*$_{\text{hapto}}$ $p = 0.050$, $r^2 = 0.12$). This difference is likely inherent in the different algorithms used to estimate PIC from each satellite (see following paragraph). Nevertheless, these correlations demonstrate correspondence between Chl *a*$_{\text{hapto}}$ measurements and satellite PIC, both of which relate to relative coccolithophore abundance.

Two radiance-based PIC algorithms can be used to relate water-leaving radiance to calcite absorption and scattering properties: a two-band algorithm (Balch et al., 2005) and a three-band algorithm (Gordon et al., 2001). The North Atlantic subtropical gyre exhibits relatively low PIC concentrations year-round. During the SeaWiFS/MODIS overlap period (2003–2007), PIC estimated from the two satellites revealed stark differences (Fig. 5c), possibly explained by the differences in algorithm performance in this region (i.e., sensitivity to low/background PIC concentrations). The low correspondence between the two estimates of PIC prevented the generation of a single, merged PIC time-series. We therefore report trends in PIC separately over the respective satellite eras (Fig. 8).

Chl *a* measured at BATS (30 m integral) and Chl *a* derived from satellite were significantly correlated ($p < 0.01$, $r^2 = 0.16$). In this case, Chl *a* measured by satellite displayed good correspondence between the two satellite eras and could be merged into one time-series. Following the regression technique of Brown and Arrigo (2012), we generated one continuous record of Chl *a* from 1998 to 2014 by applying linear regression over the 2003–2007 SeaWiFS/MODIS overlap period to predict these variables from 2008 to 2014 (Fig. 6c).

3.6 Regional trends in satellite PIC and Chl *a*

Linear trends in PIC derived from satellite observations are positive for most of the North Atlantic subtropical gyre (Fig. 8). Nearly all significant trends ($p < 0.05$) in PIC concentration are positive, especially during the MODIS era (Fig. 8c, d; 1998–2007 for SeaWiFS and

2003–2012 for MODIS). However, unlike at the BATS site, Chl *a* does not appear to be increasing in most of the gyre (Fig. 9). There are slight positive trends in Chl *a* around the BATS region (Fig. 9a), but these are not statistically significant (Fig. 9b). Indeed, most of the North Atlantic subtropical gyre shows a slight negative trend in Chl *a* or no trend at all. A trend of a subset of the satellite Chl *a* from 1998 to 2012 shows a slight, but nonsignificant, upward trend in Chl *a* in the BATS region (Fig. 1; 0.0009 $\mathrm{mg\,m^{-3}\,yr^{-1}}$, $p > 0.05$), just as the corresponding gridbox for Chl *a* at BATS in Fig. 7.

4 Discussion

4.1 Phytoplankton dynamics at BATS

In this study, we observed that coccolithophore populations, based on pigment data for haptophytes, are increasing at BATS and are positively correlated with DIC and $\mathrm{HCO_3^-}$ (Figs. 1b, 3, 7a). We observed opposite correlations for DIC and $\mathrm{HCO_3^-}$ with *Synechococcus*, the other major member of the phytoplankton community at BATS. Some studies have suggested that photosynthesis and growth of the coccolithophore, *E. huxleyi*, is carbon limited and could possibly benefit from increasing $\mathrm{CO_2}$ (Riebesell, 2004; Rost et al., 2003; Bach et al., 2013). Since $\mathrm{CO_{2(aq)}}$ and $\mathrm{HCO_3^-}$ concentrations increase with increasing DIC/ocean acidification, both photosynthesis and calcification could be stimulated in coccolithophores, which primarily use $\mathrm{CO_2}$ for photosynthesis and $\mathrm{HCO_3^-}$ for calcification (Bach et al., 2013). The results presented in this study support the hypothesis that coccolithophores are responding positively to increasing carbon availability, perhaps increasing their competitive ability in oligotrophic settings such as BATS. However, a threshold $\mathrm{H^+}$ ion concentration could be reached with further ocean acidification, eventually constraining coccolithophore growth.

Synechococcus, on the other hand, was negatively correlated with increasing carbon in the upper mixed layer and positively correlated with temperature. In laboratory experiments, *Synechococcus* showed only a slight, non-significant increase in growth rate under elevated

CO_2 conditions, but increased growth 2.3 fold with increasing CO_2 and temperature (Fu et al., 2007). Sea surface temperature in the upper 30 m at BATS has not increased significantly over the time period of this study (0.04 °C yr^{-1} trend, $p = 0.22$). However, positive temperature anomalies were recorded during the 2000–2004 period, a period of increased *Synechococcus* pigment abundance (and low haptophyte pigment abundance). Conditions that could favor *Synechococcus* may eventually arise with further warming, increasing the competitive ability of *Synechococcus*.

In order to examine redundancy in our correlations, we performed multiple linear regressions between Chl a_{hapto} and several of the driver variables with which it showed the strongest correlations (not shown). When DIC and HCO_3^- were regressed together with Chl a_{hapto}, all the statistical power of DIC was removed, indicating that HCO_3^- is the primary driver of the two for Chl a_{hapto} variability. Further, when temperature and NO_3^- were regressed together with Chl a_{hapto}, the statistical effect of temperature was removed. This indicates that temperature is not a controlling factor for coccolithophore variability, but rather is a proxy for nutrient concentration in relation to coccolithophore growth. A multiple linear regression of Chl a_{hapto} with both HCO_3^- and NO_3^- explained $> 50\,\%$ of the variance in Chl a_{hapto} ($r^2 = 0.52$). Other factors, such as competition or grazing, could perhaps account for some of the remaining variability. Both HCO_3^- and NO_3^- have increased significantly over this time period ($p < 0.001$). However, NO_3^- measurements are highly variable and near zero, making their accuracy questionable, and the trend is only significant if the last two years are included in the time-series (~~there were particularly high NO_3^- measurements during 2011 and 2012~~Figure S2). This is in contrast to HCO_3^-, which shows largely positive trends over this time period matching quite well with those of Chl a_{hapto} (Fig. 7).

The positive trend in HCO_3^- concentration in the upper mixed layer of the water column at BATS is most likely due to increasing absorption of anthropogenic CO_2 from the atmosphere. The upper 30 m of the water column is particularly inundated with anthropogenic CO_2 in the North Atlantic (Sabine et al., 2004; Bates et al., 2012). From 1991 to 2012, DIC concentration in the upper 30 m at BATS increased by a rate of 1.4 µmol $\mathrm{kg}^{-1}\,\mathrm{yr}^{-1}$, which is roughly the expected rate of increase given the rise in atmospheric CO_2 (see Chapter 10 in

Sarmiento and Gruber, 2006). Increasing inorganic carbon supply could also be accompanied by warmer sea surface temperatures, increased stratification, and decreased nutrient supply over the next century (Cabré et al., 2015). Enriched coccolithophore growth by this additional carbon, as well as other predicted oceanic changes with global warming, could lead to shifts in phytoplankton community structure at BATS.

Coccolithophores are not the only phytoplankton that may be responding positively to additional inorganic carbon. *Trichodesmium*, the filamentous N_2-fixing cyanobacteria, has been shown to increase growth and N_2 fixation under increasing CO_2 (Hutchins et al., 2007), yet other drivers such as sea surface temperature, nutrients, and species diversity tend to exert more control on their growth in situ (Snow et al., 2015; Gradoville et al., 2014). *Trichodesmium* has been reported to be a common component of the phytoplankton assemblages in the subtropical North Atlantic (Carpenter et al., 2004; Agawin et al., 2013; Orcutt et al., 2001), but was not specifically resolved in this study. *Trichodesmium* contain a similar suite of pigments as *Synechococcus* (Carpenter et al., 1993; Andersen et al., 1996), and therefore could be included in the Chl a_{syn} fraction of our calculations. This would aid to explain the negative correlation between Chl a_{syn} and NO_3^- (Fig. 3), and further explain why Chl a_{syn} is more abundant than Chl a_{hapto} in the upper water column during warmer, more stratified periods.

Whatever the exact components of Chl a_{syn}, Chl a_{hapto} generally shows opposing abundance with this group. We hypothesize that when the *Synechococcus* component (∼40% of Chl *a* on average at BATS) is abundant (perhaps due to a positive temperature anomaly), photosynthesis accompanying an increase in *Synechoccocus* draws down DIC (thus, HCO_3^-). Low DIC ~~provokes~~ could provoke carbon limitation of the coccolithophore population (Riebesell, 2004; Rost et al., 2003), hindering their competitive ability. Overall, however, the increase in DIC experienced at BATS during the past two decades could lead to slightly increased growth rates of coccolithophores (Riebesell, 2004; Rost et al., 2003) , perhaps bolstering their competitive ability within the phytoplankton community. If coccolithophores are becoming more competitive at BATS due to a lessening of carbon limitation, then they could continue to exert greater competitive stress on *Synechococcus*, which appear to be competing with coccolithophores for a similar niche. However, if the surface waters continue to warm in this region of the Atlantic, as predicted (Cabré et al., 2015), then *Synechococcus* could regain its competitive edge. Furthermore, ~~declining~~ increasing H^+ ion concentrations could eventually constrain coccolithophore growth (Bach et al., 2013, 2015). Small changes in DIC, nutrients, pH, or temperature could, in combination, influence which phytoplankton group (*Synechococcus* or haptophytes) dominate at any given time at BATS.

The *Prochlorococcus* group, designated by Chl a_{pro}, shows similar correlations with oceanic driver variables as for Chl a_{hapto} (Fig. 3). *Prochlorococcus* have been shown to lack a $CO_{2(aq)}$ uptake mechanism and therefore rely on HCO_3^- uptake for photosynthesis (Badger and Price, 2003), possibly explaining similar behavior to coccolithophores (also positively correlated with HCO_3^-), which use HCO_3^- for calcification. *Prochlorococcus* reside mainly in the deep chlorophyll maximum, comprising a rather small portion of the Chl *a* in the upper 30 m at BATS (Figs. 2, 4). This could be, however, due to the relatively high Chl *b* to Chl *a* ratio used in our pigment calculations (Letelier et al., 1993), which is more representative of low-light *Prochlorococcus* (Partensky et al., 1999). Yet, since ∼ 2005, Chl a_{pro} has been more common in the upper 30 m, resulting in an overall positive trend in Chl a_{pro} over the entire time-series ($p < 0.001$).

Consistent with colder, high nutrient environments in which diatoms are normally found, Chl a_{diatoms} showed a strong positive correlation with NO_3^- and a negative correlation with temperature (Fig. 3). If predicted trends in sea surface temperature and nutrient supply with further stratification are realized, then diatoms could become a reduced component of the phytoplankton assemblage at BATS (Cabré et al., 2015). Combining fine scale phytoplankton dynamics from BATS with the satellite record can help to elucidate what changes are occurring over large spatial scales.

4.2 Trends Chl *a* and Chl a_{hapto} in the subtropical gyre

Unlike at BATS, Chl *a* in the North Atlantic subtropical gyre is not increasing (Fig. 9). PIC, on the other hand, shows mainly positive trends over the whole gyre, in agreement with data from BATS – Chl a_{hapto} shows overall increasing trends, despite some periods of low abundance. Together with an absence of Chl *a* trend, this implies that coccolithophores are increasing in abundance relative to other types of phytoplankton in the subtropical gyre. Accompanying this conclusion, however, are uncertainties associated with the satellite-derived PIC estimates (see Section 4.3 on Limitations of this Study). Even so, ratios relating Chl a_{hapto} to PIC can further elucidate confidence in satellite-derived PIC estimates.

Data on the amount of Chl *a* per coccolithophore cell allows the calculation of cell concentration of coccolithophores in the surface waters at BATS. Using a value of 0.26 pg Chl *a* per cell (Haxo, 1985), mean coccolithophore cell concentration in the upper 30 m is 143×10^3 cells L^{-1} (corresponding to the mean value of 37.3 µg Chl a_{hapto} m^{-3} in the upper 30 m). Employing a ratio of PIC to coccolith of 0.26 (Balch et al., 1992) and considering 15 coccoliths per cell (a minimal monolayer of coccoliths covering the cell) under nutrient replete conditions and 100 coccoliths per cell under severe nutrient limitation (Paasche, 1998), we arrive at a PIC concentration range of 0.56 to 3.73 mg PIC m^{-3}. This range corresponds well to the average satellite-derived PIC concentration in the BATS region (Fig. 1a) over the study period: 2.71 mg PIC m^{-3} for SeaWiFS (standard deviation = 0.50) and 2.66 mg PIC m^{-3} (standard deviation = 0.39) for MODIS. The relatively high satellite-derived PIC concentration further suggests that coccolithophores may be experiencing nutrient limitation at BATS (and producing additional coccoliths in response; Paasche, 1998). Therefore, even given the multiple sources of error involved with satellite-derived PIC estimates and pigment analyses (see below), we feel the strong predominance of positive trends in PIC, along with the BATS Chl a_{hapto} data, suggests that coccolithophores are proliferating in this region (Fig. 8).

4.3 Limitations of this study

There are several caveats of this study that must be discussed before these results can be put into context. First, a primary assumption in this paper is that the haptophyte group is mainly composed of coccolithophores. Though high haptophyte diversity has been reported in open ocean regimes (Liu et al., 2009), this does not necessarily contradict the assumption that coccolithophores are the dominant type of haptophyte. ~~Furthermore, other studies have~~

~~shown~~ While several studies describe coccolithophores to be the dominant haptophyte ~~in open ocean sites, such as BATS (see Methods; Dandonneau et al., 2006; Lomas and Bates, 2004~~ ~~We therefore assume~~ at BATS (see Methods; Lomas and Bates, 2004; Steinberg et al., 2001) , this claim remains unsupported by specific data. We use the significant positive correlation between Chl a_{hapto} and cell counts from Haidar and Thierstein (2001) to support our assumption that Chl a_{hapto} is mainly representative of coccolithophores (Figure S1). However, as with any study that derives phytoplankton community composition from signature pigments, inherent uncertainties are associated with changes in pigment content within a phytoplankton group over time.

Overall pigment and PIC concentration per coccolithophore cell may be influenced by environmental conditions. For example, photo-acclimation and nutrient limitation can invoke changes in pigment composition or calcification that are not necessarily associated with changes in overall abundance (Behrenfeld et al., 2015; Paasche, 1998). Dominant species shifts within a phytoplankton group could also influence pigment and/or PIC measurements. Nevertheless, given upward trends observed for both Chl a_{hapto} and PIC (Figs. 5, 8) we feel the most probable explanation of these observations is increases in overall coccolithophore abundance. However, satellite-derived PIC measurements also contain inherent uncertainties.

Radiance-based algorithms for deriving PIC from satellite reflectance data are formulated to capture the light scattering properties of the numerically dominant coccolithophore, *E. huxleyi*, but also capture detached or detrital coccoliths (Gordon et al., 2001; Balch et al., 2005). PIC concentrations in the North Atlantic subtropical gyre are comparatively low, generally ~2.7 mg m^{-3}, compared to other coccolithophore bloom regions, which have PIC concentrations between 10 and 100 mg m^{-3} (Balch et al., 2005). The low concentrations of PIC observed in the North Atlantic subtropical gyre could be within background error or nearing the sensitivity threshold of the instrument. Errors in satellite-derived PIC can arise from atmospheric correction, inclusion of other suspended minerals (such as silica; "opal contamination"), and/or the influence of chlorophyll or colored dissolved organic matter (see Balch et al., 2005). However, these errors can be minimized by binning in space and time, as we have done in this study (using monthly, 9 km data rather than daily, 4 km data). It is also curious that SeaWiFS-derived PIC data better matches the Chl a_{hapto} estimates from BATS than the MODIS PIC (Fig. 5). On one hand, this may be indicative that other 19'-hex-containing haptophytes were responsible for the increase in Chl a_{hapto} at BATS during the last several years of the time-series. On the other hand, the predominance of upward trends in MODIS-derived PIC for areas around BATS (Fig. 8b,d) suggests increases in calcifying haptophytes (coccolithophores). It should be noted that MODIS PIC from the BATS region (see Fig. 1) still shows a significant correlation with Chl a_{hapto} (see Results, section 3.5).

Finally, we are limited in our trend analysis by the length of the time-series data. Figure 7 demonstrates that different start and end years can influence the sign and magnitude of our trends in, e.g., Chl a_{hapto}. In this study, we report trends in pigments from 1990 to 2012 and trends in PIC from 1998 to 2014, both of which, when employing the full time-series of data, imply increases in coccolithophore populations in the subtropical North Atlantic.

4.4 Trends in coccolithophore abundance: present and future

This is not the only study to suggest that coccolithophores are increasing in abundance in the North Atlantic. Further, using Continuous Plankton Recorder ship measurements, Rivero-Calle and co-authors (2015) document an increase in coccolithophore occurrence from ~ 2 to $> 20\,\%$ in the North Atlantic from 1965 to 2010, which they attribute to increasing $\mathrm{CO_2}$ concentrations. The data region in their study extends from ~ 40 to $\sim 65°$ N (subpolar gyre), just north of the subtropical gyre region focused on in this study. Thus, these studies combined add robustness to the conclusion that coccolithophores in the North Atlantic are increasing in abundance and are likely stimulated by additional carbon from anthropogenic sources.

Ocean acidification may eventually hinder the growth and calcification of coccolithophores, however. Recently, Bach (2015) and Bach et al. (2015) introduced the "substrate–inhibitor" ratio, describing the dependence of calcification on $\mathrm{HCO_3^-}$ (the substrate) and $\mathrm{H^+}$ (the inhibitor) concentrations. When this ratio falls below a critical level (i.e., intercellular to extracellular $\mathrm{H^+}$ concentration ratio too low) then coccolithophore calcification will be hindered, unless they evolve a mechanism for coping with low pH (Bach et al., 2015). Bach et al. (2013) demonstrate that pH starts to have a negative impact below pH$_T$7.7, whereas the BATS average pH$_T$ is ~ 8.1. Thus, critical pH levels will not likely happen for several thousand years (Bach et al., 2015). Other factors besides carbonate chemistry, such as light availability, temperature, and nutrients, likely influence coccolithophore growth in present day oceans.

4.5 Potential implications

Increases in coccolithophore abundance in the North Atlantic could have far-reaching ecological, biogeochemical, and climate effects. A shift in phytoplankton community structure could change trophic dynamics, ultimately resulting in ecosystem shifts (Pörtner et al., 2014). For example, though the evolutionary purpose of coccolithophore shells is unclear, some studies speculate they could protect against grazing (see chapter on functions of coccoliths in Winter and Siesser, 1994). Shifts to relatively more coccolithophores in a phytoplankton assemblage could reduce trophic energy available for grazers. Coccolithophore shells also function as a ballast material, sinking faster due to increased weight of the $CaCO_3$ shell, and sequestering organic matter in the deep ocean (Sarmiento and Gruber, 2006; for a recent study of export at BATS, see Lomas et al., 2010). Increases in coccolithophore abundance may have a positive impact on export production, thus a negative feedback on increasing atmospheric CO_2. In addition to bringing carbon to the deep ocean, coccolithophores produce the marine trace gas dimethyl sulfide (DMS; Keller, 1989), which affects cloud formation and climate. Therefore, overall increases in coccolithophore abundance could increase marine DMS production. Furthermore, DMS production by the coccolithophore, *E. huxeyi*, has been shown to increase with increasing temperature and ambient CO_2 (Arnold et al., 2013). Thus, changes in coccolithophore abundance could have a multitude of effects on marine ecosystems in the North Atlantic, as well as global carbon cycling and climate. These effects could be further amplified if other ocean basins show similar shifts in phytoplankton composition.

In the subtropical North Atlantic, the upper mixed layer contains particularly high levels of anthropogenic CO_2 (Sabine et al., 2004; Bates et al., 2012). We speculate that this rise in DIC is contributing to the increases in coccolithophore pigments and PIC documented in this study. However, it is not clear if phytoplankton communities in other similar oceanic ecosystems, e.g., the North Pacific subtropical gyre, will show similar changes as atmospheric CO_2 concentrations continue to increase and inundate the upper mixed layer. The aforementioned ecosystem and carbon cycle effects of coccolithophore increases could become even more prevalent in the world's ocean, or, alternatively, coccolithophore growth could be further modulated by temperature, nutrients, and light. In any case, monitoring the response of natural coccolithophore populations to increasing DIC/ocean acidification is essential for understanding effects of anthropogenic carbon emissions on the world's oceans.

5 Conclusions

In this study, we showed that, based on pigment and satellite-derived PIC measurements, coccolithophores appear to be increasing abundance in the subtropical North Atlantic. Coccolithophores seem to be responding positively to additional inorganic carbon in the upper mixed later of water column, but are also correlated with NO_3^-. These results complement those of Rivero-Calle et al. (2015), who also document an increase in coccolithophore occurrence in the North Atlantic, albeit farther north, stimulated by anthropogenic CO_2 emissions. Increasing coccolithophore abundance is contrary to what numerous laboratory studies have predicted, highlighting the importance of in situ observations. Growth of coccolithophores could, however, be eventually inhibited by decreasing pH and/or other environmental effects of climate change.

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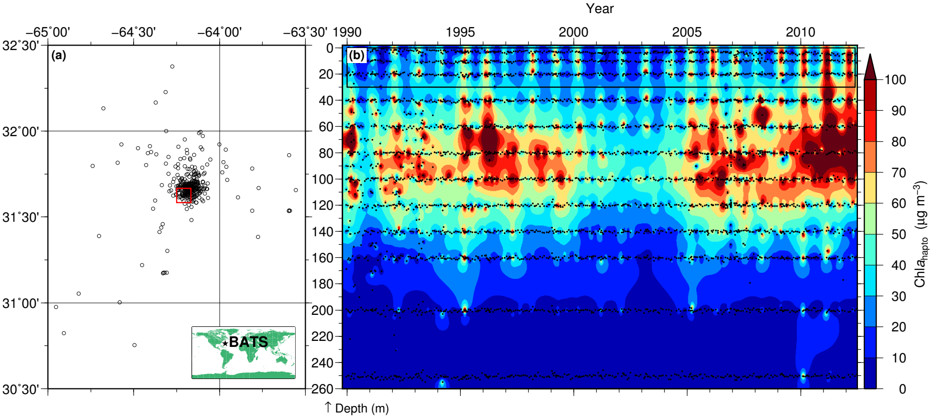

Figure 1. Horizontal **(a)** and vertical **(b)** distribution of pigment measurements taken at BATS (black dots) overlaid on a contour plot of HPLC-measured Chl a_{hapto} at BATS. The red box in **(a)** shows the PIC grid cell containing the most BATS measurements (PIC data shown in Fig. 8), while the black box in **(b)** shows the upper 30 m of measurements used for PIC-Chl a_{hapto} correlations and correlations presented in Fig. 3.

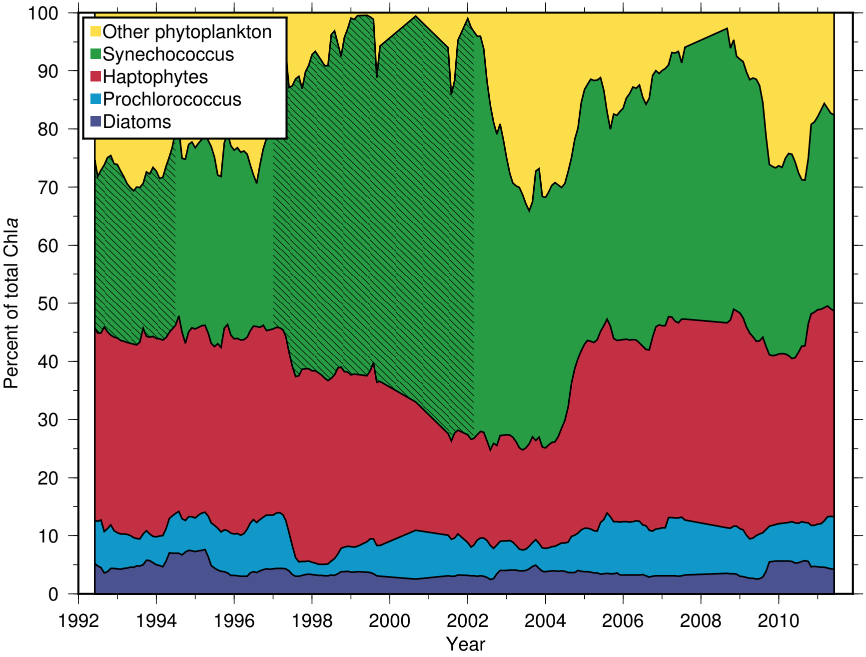

Figure 2. Percent of Chl *a* from main phytoplankton groups at BATS from 1992 to 2012 in the top 30 m of the water column derived from signature pigment and Chl a_{total} concentrations, deseasonalized with a 1 year boxcar filter (purple = diatoms, blue = *Prochlorococcus*, red = haptophytes, green = *Synechococcus*, yellow = other phytoplankton). Hatched area indicates missing pigment data for *Synechococcus*.

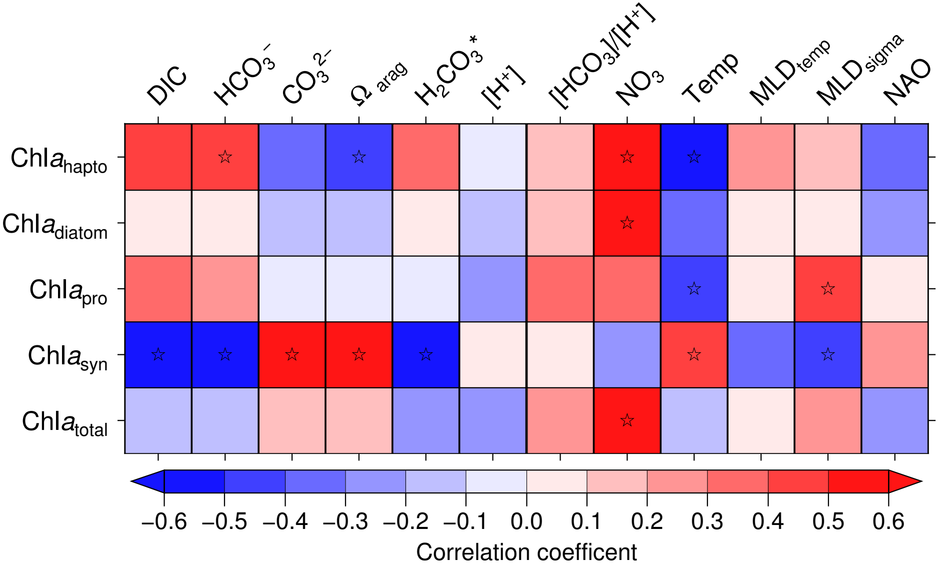

Figure 3. Correlation coefficients between Chl *a* components and various oceanographic measurements made in the upper 30 m of the water column at BATS, NAO index, calculated mixed layer depth (MLD; see Methods) and derived carbonate chemistry parameters. Stars indicate the absolute value of the correlation coefficient is greater than 0.4.

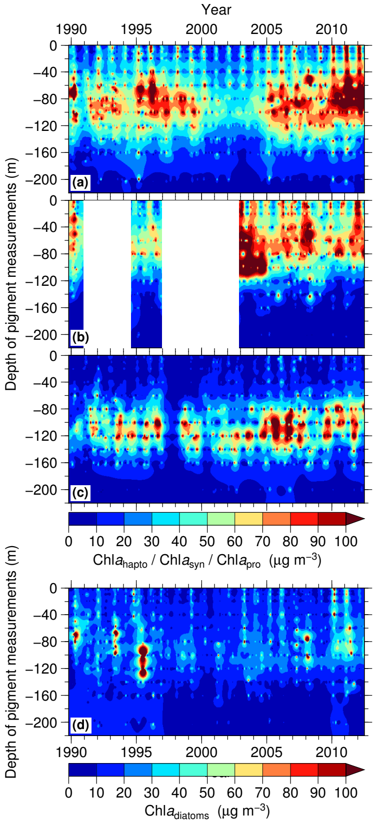

Figure 4. Temporal evolution of the vertically-resolved Chl *a* concentration from the main phytoplankton present at BATS derived from signature pigments from 1990 to 2012: **(a)** Chl a_{hapto}, **(b)** Chl a_{syn}, **(c)** Chl a_{pro}, and **(d)** Chl $a_{diatoms}$.

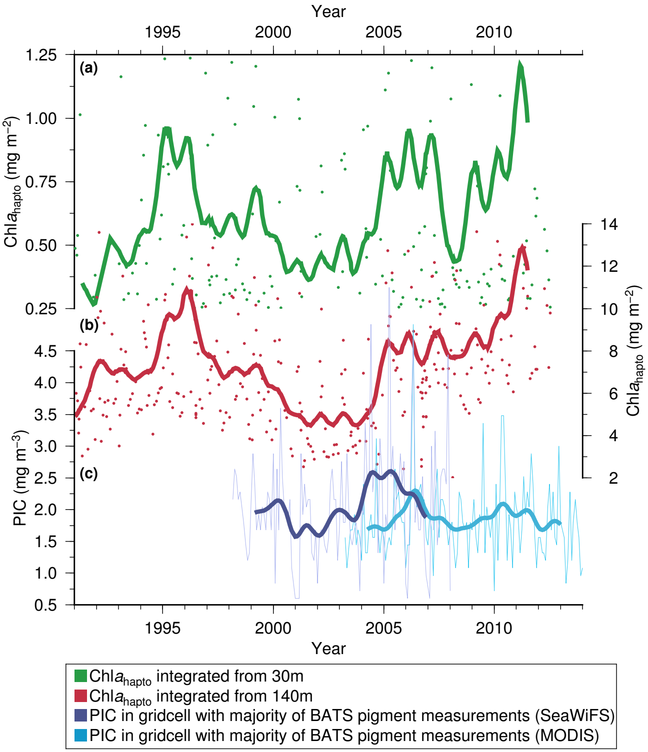

Figure 5. Chl a_{hapto} measured at BATS and satellite-derived PIC. Chl a_{hapto} integrated from 30 m is shown in **(a)**, while Chl a_{hapto} integrated from 140 m depth is shown in **(b)**. PIC data shown in **(c)** was obtained from the 5 min satellite grid cell with the most BATS measurements (see Fig. 1). Bolder lines represent a 2 year Gaussian filter on the data. We restricted the y-axes in panels (a) and (b) to highlight the filtered data; see Figure S3 to view all unfiltered data points.

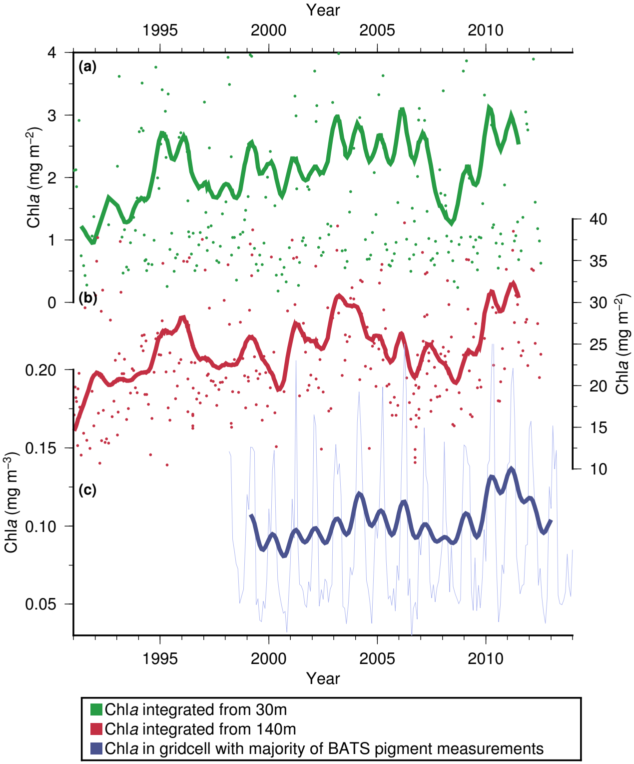

Figure 6. Chl a_{total} measured at BATS and satellite-derived Chl a. Chl a_{total} integrated from 30 m is shown in **(a)**, while Chl a_{total} integrated from 140 m depth is shown in **(b)**. Chl a data shown in **(c)** was obtained from the 5 min satellite grid cell with the most BATS measurements (see Fig. 1). Bolder lines represent a 2 year Gaussian filter on the data. We restricted the y-axes in panels (a) and (b) to highlight the filtered data; see Figure S4 to view all unfiltered data points.

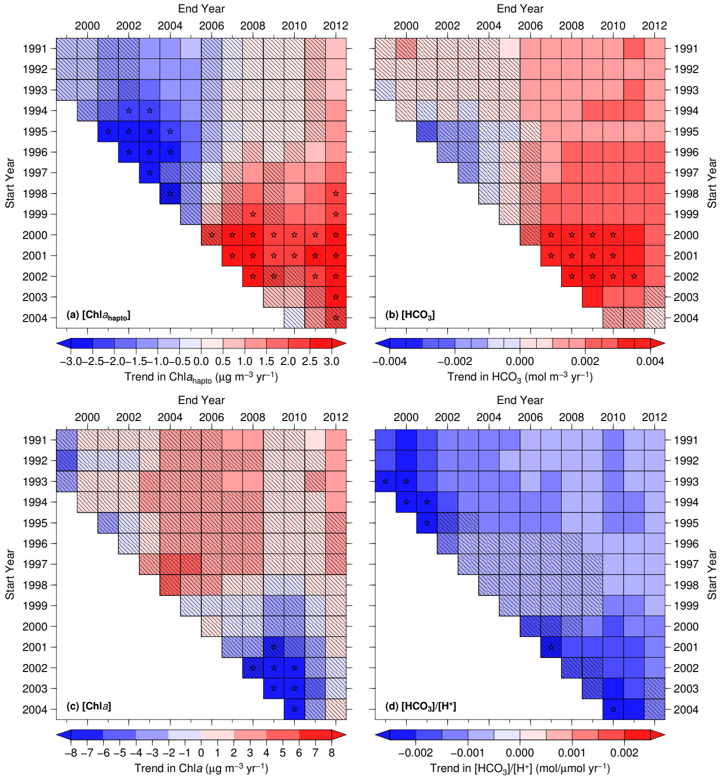

Figure 7. Linear trends for a range of start and end years in **(a)** Chl a_{hapto}, **(b)** HCO_3^-, **(c)** Chl a, and **(d)** ratio of $HCO_3 : H^+$. All trends are based on mean concentrations measured at BATS in the upper 30 m of the water column. Boxes with hatch lines demarcate nonsignificant trends. Stars indicate absolute values of trends are greater than 0.2 µg m^{-3} yr^{-1} for Chl a_{hapto}, 0.003 mol m^{-3} yr^{-1} for HCO_3^-, 6 µg m^{-3} yr^{-1} for Chl a, and 0.002 mol/µmol yr^{-1} for $[HCO_3^-]/[H^+]$.

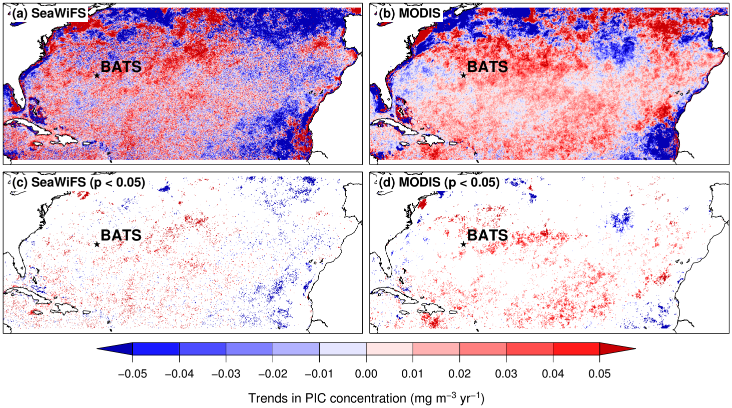

Figure 8. Trends in PIC concentration derived from satellite data for **(a)** and **(c)** SeaWiFS (1998–2007), and **(b)** and **(d)** MODIS (2003–2014). Bottom panels shows significant trends ($p < 0.05$).

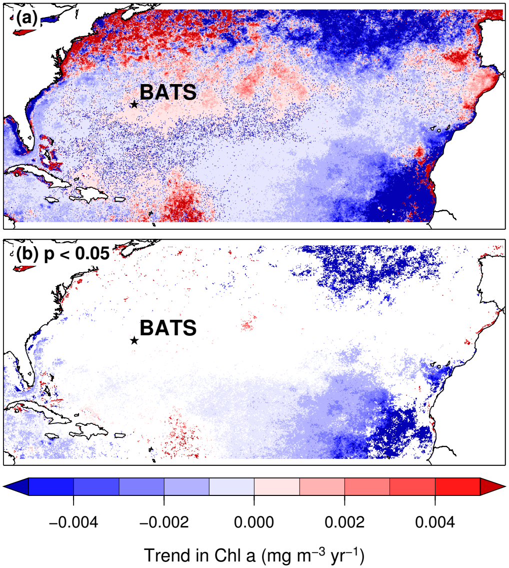

Figure 9. Trends in Chl *a* concentration derived from merged satellite records: SeaWiFS and MODIS (1998–2014). The bottom map **(b)** shows significant trends ($p < 0.05$).