Dear Editor,

Thanking you for editing the manuscript (BG-2015-240) and sending reviewers' comments. Herewith we submit a revised manuscript for consideration for publication in BG along with reply to reviewers' comments. The point to point reply to comments/suggestions made by the reviewers is presented below (in blue font, preceded by "Reply"). In the accompanying revision, changes associated with the reviewer's comments are either highlighted in yellow or noted by an embedded comment. If there are any questions, please don't hesitate to contact me.

Sincerely,

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Reviewer#1

The authors have examined the elemental stoichiometry of total organic material (TOM) and particulate organic material (POM) in the upper 100 m of the water column, as well as the inorganic nutrient pools, over an eight year period at the BATS station. The aim of this study was to quantify C:N:P ratios in all these pools and their relationship to biogeochemical cycling, community structure and the canonical Redfield ratio. The also analyzed the annual and seasonal variability in these parameters. All data were obtained from the publically available BATS web archive.

They found that the TOM C:N:P ratios exceeded those of the POM and they present linkages between the observed TOM and POM seasonal variability to that of phytoplankton cell abundance and taxonomic group, as well as potential climate drivers for the observed long-term variability in C:N:P stoichiometry.

Overall this is a rather straightforward analysis of time-series data from BATS. The C:N:P work appear solid, but I have questions about how the data were used and how that may influence the interpretation of the results. In addition, I believe some restructuring of the manuscript would help to improve its readability. For example there are quite a bit of data that is presented in the discussion section that would fit better into the results section.

Reply: We thank the reviewer for going through our manuscript thoroughly. We appreciate the comments and all the concerns. We have addressed them below one by one.

Detailed comments:

P9276, ln 16. “C:N:P ratios in the TOM pool were more than twice that in the POM pool”. I think this needs to be rephrased. The data in table 1 shows C:N and N:P being ~2× higher in TOM compared to POM, whereas C:P is ~4-5× higher in TOM than POM. I suggest breaking this out in its components to make this clearer.

Reply: We have broken the sentence into two parts as suggested by the reviewer (ll 44-46), and stated ‘at least’ rather than ‘more than’ for C:N and N:P ratios.

P9280, ln 20. At what depth were the sediment traps deployed? (this appears later in the discussion, but should be mentioned in the Materials and Methods).

Reply: Sediment traps were deployed at 200 m depth. We have now mentioned in the M&M section (ll 155-157).

P9281, ln 5. How were the ‘depth mean ratios’ calculated? Was an elemental ratio calculated for each depth and then average over the 7 depths from 5-100 m, or was an average concentration of each element calculated and then the ratio made? How do you weight average the data when the sample spacing is not even (i.e. spacing 5m, 5m, 10m, 20m, 20m, 20m and 20m)? Have you

thought about integrating the TOM and POM inventories over your sampling depths instead? This may alter the results but may be more relevant for the comparison of the two depth ranges chosen (0-25 m and 25-100 m).

Reply: We first calculated the average concentration of each element over the depth segment (e.g., 0-25 m) and then the ratios were calculated from those averages. We have specified this in the manuscript now (ll 171-173). Also, this approach does not require a 'weighting' function to be applied.

5, 10, 20, 40, 60, 80 and 100 m were the target depths but actual depths (sometimes) changed during CTD operation by a few meters (~2-3 m). So all the depth sampled above 25 m were put in 0-25 m, while below it were put in 25-100 m depth.

Our concentrations have µmol kg^{-1} units and changing units to µmol L^{-1} might propagate uncertainly due to sometimes uncalibrated salinity sensor. Moreover, our analysis is mainly based on 0-25 m depth, where samples were almost equally spaced, and the MLD was hardly shallower than 25 m so concentration of different elements was quite homogeneous. Hence, we have decided not to use integrated values, but the reviewer's comments are duly noted.

Ln 20. Was this trend in TOP based on the depth averaged concentrations over 0-100 m? It is hard to see any 'trends' in the contour plot. My impression of the plot is that 2007 had unusually low TOP whereas during 2008 TOP appeared to be unusually high. Would you get a negative trend instead if using data from early 2008 to early 2009 that would also be significant?

Reply: Yes, this trend in TOP was based on the depth averaged concentrations over 0-100 m. It is hard to see in the contour plot. We discovered it from our ratio analysis (Fig. 2) and analyzed TOP separately. TOP values were indeed low in the beginning of 2007 but increased gradually until January 2008. Early 2008 to early 2009 TOP data show negative trend over time but it is much less robust (r^2 = 0.39, p-value: 0.03) compared to 2007-2008 trend (r^2 = 0.77, p-value: <0.001).

P9282, ln 14. What determined the choice of depth division of the water column at 0-25 m and 25-100 m?

Reply: We wanted to analyze annual variation in elemental ratios in different depth segments. Segments were based on MLD, which was normally not shallower than 25 m depth during the summer stratified period. Thus we took this as a 'surface' depth segment. We were also concerned that preferential degradation of TOP should not change annual variation in elemental ratios and hence we decided to separate into 0-25 depth segment.

Ln 16. How was the 0-25 m concentrations calculated when sampling depths were 20 and 40 m? Were the data interpolated between 20 and 40 m?

Reply: As stated above, 5, 10, 20, 40, 60, 80 and 100 m were the target depths but actual depths (sometimes) changed during CTD operation by a few meters (~2-3 m). So all the depth samples above 25 m were put in 0-25 m, while below it were put in 25-100 m depth. Thus, the 20m sampling was always in the shallow segment and 40m always in the deeper segment. Because we didn't integrate the data, but rather averaged data above/below a depth cutoff, there was no need to interpolate the data.

Ln 23-25. Does Trichodesmium not contribute to POM? I do not really see a peak in TOC, but TON and PON peak in month 6. Is that what was meant? This 'peak" also is seen in the 25-100 m portion but that is not mentioned in the text. I would suggest switching the wording around..from " the occurrence of higher Trichodesmium colonies" to " the higher occurrence of Trichodesmium colonies..." or "the higher number of Trichodesmium colonies".

Reply: Trichodesmium does contribute to POM but it would hard to see the changes in POM due to the fact that they are particularly patchy in distribution and not very abundant overall so it is actually rare that whole Trichodesmium colonies are captured on the filtered and then measured as POM. However, as they release N (as DON) simultaneously as they fix N_2, we see more variation in DON (or TON for the present case) that PON because of the buildup of the former. TOC also peaks in the fifth month but remains saturated afterwards. We have mentioned the similar peak in 25-100 m portion and changed the wording as suggested (ll 214-216).

P9283, ln4-9. Much of this text is an iteration of the first paragraphs of the Results section. I would suggest moving the earlier text and incorporate that under section 3.2.2. instead. Also, see line 7-8 in discussion, which is very similar to what this paragraph is saying, but stated more clearly.

Reply: In the first paragraph of results section, we have discussed the entire time series (Fig. 2). Under the section 3.2.2, we have discussed the patterns in terms of deep mixing. However, we agree that there was some repetition so we have shortened the text to improve clarity and readability (ll 173-180)

Ln 10. "Minimal variability in concentrations and ratios in the 25-100 m depth horizon.." How was that determined? I find Figs 4 and 6 remarkably similar in terms of the range in mean concentrations, seasonal patterns and variability (error bars) in the N and P pools. The N:P ratios also look quite similar in Fig 5 and 7. Only TOC and POC seem to differ somewhat in concentration range, variability and pattern between the two. I would suggest changing "25-100 m depth horizon" to "25-100 m depth range"

Reply: Some of the trends that we have discussed were not as prominent in 25-100 m depth range as they were in 0-25 m depth range. We have discussed this in the manuscript now (e.g., ll 214-216). In addition, TOC and POC values were significantly lower in the 25-100 depth range compared to that in the 0-25 m depth range, as suggested by the reviewer. We have changed 'horizon' to 'range' (Line 226).

P 9284 – Discussion. The discussion currently contains quite a bit of new data that I believe should be better presented under the result section. E.g. the trap flux data, flow cytometry and chlorophyll.

Reply: We have added new data into the results section (added two new sections - 2.3.4 and 2.3.5; ll 238-250). The reason for not including it in the first version is that much of that data was presented as a result in Lomas et al. 2013 (overview of BATS data), but in a different context. We agree that including it here as a result is also appropriate.

P9285, ln 2-4. "On the contrary, our data suggests that TON values increase with depth while TOP values do not change (Figs 4 and 6)." From Figs 4 and 6 it does look like TOP remains fairly constant in the two depth ranges compared, whereas TON goes up a little with depth. However, the TON:TOP ratios in Fig 5 and 6 doesn't seem to reflect this very clearly, and it even looks like TON:TOP may be slightly lower on average between 25-100 m than above. Am I misinterpreting these data or are there something else I am missing?

Reply: We thank the reviewer for this observation. TON indeed goes up with depth and TON:TOP is also slightly lower at 25-100 m than above. But our interpretation for TOP was not completely correct. While comparing TOP data at these two segments, we found that it was around 5% higher in the 25-100 m than 0-25 m depth, which is difficult to see in the Figures. We have revised the sentences accordingly to make this more clear and eliminate confusion (ll 274-276).

P9286, ln 5-9. "..the gradual increase in Chlorophyll a during the four months prior to deep mixing is due to a similar increase in MLD before deep mixing". Is this to mean that the increase in chlorophyll is due to increased nutrient influx into the 0-25 m depth range? Could the annual pattern in chlorophyll a concentration be explained by the changes in light flux over the yearly cycle? I.e. phytoplankton containing more chlorophyll during the winter months with lower light flux, but not necessarily more biomass?

Reply: Winter mixing, which results in spring blooms thereafter due to nutrient injection into the euphotic zone, is a well recorded phenomenon at BATS. Light could be a limiting factor in the winter and hence the blooms occur during spring. Conceptually, as fall progresses and the MLD increases due to surface cooling, phytoplankton see on average a lower light level which is compounded by the decreasing annual light pattern. So there is likely some photoacclimation going on. This is further supported by the observations of Wallhead et al. 2014, that show that phytoplankton C does not increase, relative to summer, when the MLD is deepening and thus the Chl:C in phytoplankton is arguably increasing. Given that availability of light data is not consistent, and the assumptions involved, we have raised this as a potential explanation but do not state it as a 'conclusion'.

Ln 10-14. How were these correlations made? Depth averaged over 0-25 m, or 0-100 m? It is unclear as written. Figure 9 shows only 0-25 m data, but using only such a shallow range may

result is a skewed picture. How would data from the full euphotic zone impact the interpretation of the influence of the taxonomic groups on the C:N:P stoichiometry of POM?

Reply: Correlations were made over the depth average 0-25 m. We have mentioned in the manuscript now (Lines 309 and 312). We have checked and found Figure 9 (0-25 m depth) does not give a skewed picture. Patterns are the same in the 0-100 data but they are not as prominent as in 0-25 depth likewise for the elemental concentration parameters. Moreover, our focus is mainly in the 0-25 depth. We thank the reviewer for his/her comment but we believe the presentation and interpretation are accurate.

P9289, ln 6. "Such ratios appear to be largely driven by. . ." This sentence seems to be referring to the average C:N:P ratios of both TOM and POM. Was that the intent? Or was it supposed to refer to the annual or seasonal variability observed, or the out of Redfield ratio that can be inferred from the Synechococcus and Prochlorococcus? I suggest adding some words to make the sentence clearer.

Reply: We meant that the seasonal variation in POM stoichiometry appears to be largely driven by the growth of Synechococcus during winter mixing. The Redfield ratio in POM can be explained by Prochlorococcus abundance. We have made both of these statements more clear now (ll 383-387).

Table 1. What is the rational behind the presentation of data collected prior to this study's window for some parameters? What criteria was used to create the ratios? (The number of observations are much reduced for the ratios relative to each parameter measured by itself).

Reply: More data provide better statistics so we wanted to put all the BATS data on the parameters we have analyzed in the Table 1. But for our deep mixing analysis, it was fair to use only concurrent data. Ratios were calculated for each depth, where both (POM and TOM) the parameters were measured. In many cases, both parameters were not measured at the same depth and hence the number of observations are much reduced for the ratios relative to each parameter measured by itself.

Figs 4-7. (see above question for ratios in Table 1). Are the ratios derived from a different subset of samples than what is presented for each parameter measured by itself? There are no "n" number mentioned in the figure legends.

Reply: These ratios are derived from a subset of the data listed in the Table 1. However, here we first estimated average concentrations of each element over the depth segment (e.g., 0-25 m) and then the ratios were calculated (please see first comment). We have specified this in the manuscript now (ll 171-173). This way, we could include all the data for the time segment January 2005 - December 2011. Now one bar in each figure is obtained from the seven data points (one each year from 2005 – 2011). But this one (of those seven data) datum is estimated

from around three points (5, 10, 20 m targeted depth). Hence, mentioning “n” in the figures could be confusing, but we have attempted to make it clearer in the text (ll 171-173).

Minor:

“Redfield Ratio” or “Redfield ratio”. Both are used throughout. I suggest using only one version.

Reply: We have corrected it throughout the manuscript to “Redfield Ratio”.

P 9286, ln 7. Spelling Chlorophyll

Reply: Corrected (Line 305).

Suggestion on Figs 4-9. Box plots would be a very nice way to present these type of data as the data sets are large and the box plot format gives so much more information than the mean and std-deviation.

Reply: We welcome this suggestion. We present our data in box plots for the Figures 4-9.

Reviewer#2

Singh et al. use suspended particulate organic matter (POM) and total organic matter (TOM) from the upper 100m, as well as exported POM between 100-500m from the BATS database to investigate ecosystem elemental stoichiometry (C:N:P). They find the C:N ratios in the particulate pools approximate Redfield proportions but that ratios relative to P are much higher than Redfield (i.e. C:P and N:P in both the total and particulate pools). They link these higher than Redfield elemental ratios to plankton abundance, primarily the cyanobacteria Synechococcus and Prochlorococcus and to a lesser extent pico- and nanoplankton. They also suggest elemental ratios differ as a function of growth rates and that elemental stoichiometry is related to the Arctic Oscillation.

Overall I am supportive of this manuscript. It is a good set of data that lends strong support for a non-Redfieldian ocean. While I think this view is becoming widely accepted among oceanographers, showing it in the BATS database is nice in that this data set is used by so many for modeling that part of the ocean. Assuming Redfield proportions in an ecosystem or biogeochemical model based on BATS data is not really an option as shown by this paper. However, the manuscript is not yet ready for publication. I have several comments/questions for the authors that I believe need to be addressed prior to publication.

Reply: We thank the reviewer for comments and all the concerns, and their support for the value of the paper. We have addressed them below one by one.

1. line 63-66, and again at lines 360-364, here the authors claim there is a lot of support for proximate P limitation of productivity in the waters at the BATS site. They then cite several papers of which I would argue none actually support P limitation of productivity. The Lomas et al. 2010 paper actually uses the term P stressed instead of limitation and argues growth of the phytoplankton is Redfieldian when DOP is taken into account. The other papers cited assume P limitation based on Redfield N:P or C:P stoichiometry (i.e. if ratios are greater than 16 or 106 respectively than PO_4^{3-} is limiting). However, this cannot be the case if the primary producers themselves are not Redfieldian (i.e. if their ratios are naturally greater than Redfield proportions). The Bertilsson et al. and Heldal et al. papers show that even under nutrient replete conditions the cyanobacteria have N:P and C:P ratios higher than Redfield. If this is the case one cannot assume proximal P limitation based on higher than Redfield stoichiometry.

Reply: We completely agree with the reviewer; assessing 'limitation' is very difficult. There are also other studies that suggest N is the proximal limiting nutrient in this part of the ocean. At some level it depends what your response variable is, e.g., growth rate vs. chla content, etc. We have now clearly stated throughout that the North Atlantic is potentially P stressed (ll 64-66, 368-370, 372-374).

2. Related to the above is that the assumption of P limitation could then be assumed if the particulate ratios were greater than the nutrient replete ratios of the cyanobacteria which in the

BATS data they seem to be (though not by a lot). However, Singh et al. state that phytoplankton account for only 25% of the particulate matter. What is the other 75%? If only 25% of the particulate matter is phytoplankton than it is difficult from the presented data to know their elemental ratios and thus whether or not they are > or < the nutrient replete stoichiometry of the cells.

Reply: We have estimated that *Prochlorococcus*, *Synechococcus* and *Picoeukaryote* contribute up to 75% to the PON. Other phytoplankton (e.g., diatoms, dinoflagellates, *Nanoeukaryotes*), diazotrophs (e.g., *Trichodesmium*), bacteria and zooplankton (both micro and macro) might contribute to the other 25%. *Trichodesmium*, which is abundant during summer at the BATS, has an N:P ratio that varies from 42 to 125 (Karl et al., 1992). But we do not have elemental content of these other (25%) plankton so we cannot state this in the manuscript.

I would argue there is little direct evidence for P limitation of productivity in these waters and that elemental ratios, in this system where phytoplankton are only 25% of the particulate pool, cannot be used to determine limitation status of the primary producers. There is a lot of evidence that shows adding N to the waters of the North Atlantic Subtropical Gyre stimulates primary productivity (see the Moore et al. 2013 review paper which the authors cite). There is evidence also that shows adding PO_4^{3-} to the same waters does not stimulate primary productivity. Additionally, the term PO_4^{3-} limitation (end of paper) should not be used, instead use P limitation as at the start of paper.

Reply: We agree with the reviewer, please see response to comment 1 above. We have now stated that the North Atlantic is P stressed (ll 64-66, 368-370, 372-374).

3. line 51- add vary between ratios and with

Reply: added (line 52).

4. 2nd to last sentence of abstract- sentences like this are vague. They do not say much really and do not add to the manuscript. It is better to state what the climate variability – C:N:P relationship is and means. The authors should examine the manuscript throughout and clean up these types of vague sentences or get rid of them.

Reply: We have revised such vague sentences throughout the manuscript (e.g., ll 53-54).

5. Line 154- change 2nd as to and

Reply: Changed (line 154)

6. Line 190 end of first sentence- cite figure? Fig. 2? Make sure Figures and panels are cited throughout manuscript.

Reply: Cited throughout the manuscript (line 190).

7. Line 205 -206, why not order your figures in the same order as they are presented in the results. So Fig. 5 and 6 would be switched so this sentence cites Fig. 4 & 5. It is easier for the reader to just jump to the next figure as they read than to have to jump ahead 2 figures and then back.

Reply: We have changed the order as suggested by the reviewer. Figs. 4 and 5 compare elemental concentrations at 0-25 m and 25-100 m depth range, while Figs. 6 and 7 compare elemental stoichiometry at 0-25 m and 25-100 m depth range.

8. Line 206-215- Figure 6 is cited here but is not really presented or compared to figure 4. It makes sense to present them together and the differences or similarities between the pools at each depth range.

Reply: We have discussed Fig. 6 (now Fig. 5) in detail now in connection with figure 4 (e.g., ll 215-217). Please see prior comment and response as well.

9. Line 214-215- seems like POP followed same trend, and TOP increased with mixing

and remained high and variable until the next season.

Reply: We agree. We have revised the sentences accordingly to make this observation more clear (ll 214-215).

10. Line 224- It would be good to actually compare variability- is the variability really that different? For some things yes– e.g. TOC:TON for others maybe not PON:POP. Also Fig 5 c legend reads TOC:DON not TOC:TON

Reply: Yes, variability in the two depth ranges were significantly different. We have made these clarifications and comments throughout where appropriate. We have corrected the figure caption.

11. Line 235- do you really think biological uptake between 100-500 is responsible? What uptake is this- heterotrophic? More detail please

Reply: That was a mistake in interpretation on our part. We have deleted that section of the paper. However, depending upon how you define the euphotic zone it may extent to 150m in the Sargasso Sea. Indeed we can see living phytoplankton that deep or deeper and so while surely from say 200-500m is drive by heterotrophic activity, 100-200m remains part of the transition zone from net particle production to net particle consumption.

12. The results end without presenting the flux data, instead it is at line 249 in the discussion. It should be in the results. Also the relationship to the AO is not presented in results- why is that?

Reply: We have presented the P flux data in the results section now; this was also a comment of the first reviewer (ll 239-245). The relationship to the AO was not presented in the results simply because the results were the presentation of the stoichiometric data and the link to AO was a derived 'outcome' of the discussion when trying to discuss and interpret patterns. So we feel it is appropriate to leave mention of the AO in the discussion.

13. Line 250 refers to POP flux but cites Fig 8A & B, 8A is PON flux.

Reply: We have corrected this (Line 244).

14. Line 255- change also almost to more than

Reply: Corrected (Line 268)

15. Line 257- delete however (it is not appropriate in this sentence).

Reply: Deleted.

16. Line 263-264- are these differences significant

Reply: This sentence has been modified based on the comments from Reviewer 1 (ll 275-277).

17. Line 264-268- this again is not a very convincing sentence just a statement of importance that is speculative. I think you need to point out how the data is important. I am not sure how the data you have supports DOM sustaining phytoplankton growth. Something more detailed as to how this data supports this is requested.

Reply: We have revised the sentences and substantiated our claims more soundly (ll 277-282). We hope the reviewer finds the new text satisfactory.

18. Line 273- do the changes in POM account for the changes in TOM or do there have to be DOM changes?

Reply: POM contributes ~10% to TOM (comparing POM and TOM concentration in the Fig. 4) and it is unlikely to account for changes in TOM (looking at the seasonal changes upto 5 µmol kg^{-1} in the TOM, Fig. 4). So we believe that they have to be predominantly due to DOM changes.

19. Line 305- at the BATS site.

Reply: Corrected (Line 319).

20. Line 306-307- why a mixture? N:P of Pro and Syn is same- could be a mix or could be either. Suggests cyano influence on PON:POP.

Reply: Yes, it could also be either theoretically. We have observed *Prochlorococcus* and *Synechococcus* both so we state it as mixture. That said we have clarified this sentence.

21. Line 313-314- this sentence refers to ratios, but the figure does not have ratios.

Reply: We have moved the figure reference to next sentence, where it was more appropriate and refers to the correct figure (line 332).

22. Line 320-321- fine hypothesis- but does it make sense? phytoplankton make up ~25% of the POM (15% of that is SYN) plus some Pro and Picos. So less than 10% can be nanos- if they require low P would the changes you see in their abundances alter the TOP concentrations to the extent you see?

Reply: As we have stated above, *Prochlorococcus, Synechococcus* and *Picoeukaryote* contribute up to 25% to the PON (contribution of all phytoplankton community to POM would be much higher). We do not know the contribution of *Nanoeukaryotes* to POM (which might be less than 10%) so we would like to keep our hypothesis as such. That said, we recognize the reviewers comment and have tried to expand it such that readers can better understand the context and our point of view on the hypothesis.

23. Line 331- did you do correlation analysis? If so shouldn't you report r not r^2.

Reply: Yes, we did correlation analysis, and thus now report the value as 'r'.

24. Lines 335-339- I am not sure how dilution of the inorganic pools affects the ratios of the organic pools? Some more detailed explanation is requested.

Reply: That was an incorrect formulation. We found that the mixing is too complex a process to explain ratios of the organic pools, hence we have deleted that part of the text.

25. Line 360-364- I do not see how this paragraph fits in this section relating to microbial export. Seems out of place. Plus see comment 1 in reference to this paragraph.

Reply: We agree. We have deleted this part.

C:N:P stoichiometry at the Bermuda Atlantic Time-series station in the North Atlantic Ocean

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Abstract

Nitrogen (N) and phosphorus (P) availability, in addition to other macro-and micronutrients, determine the strength of the ocean's carbon (C) uptake, and variation in the N:P ratio of inorganic nutrient pools is key to phytoplankton growth. A similarity between C:N:P ratios in the plankton biomass and deep-water nutrients was observed by Alfred C. Redfield around 80 years ago and suggested that biological processes in the surface ocean controlled deep ocean chemistry. Recent studies have emphasized the role of inorganic N:P ratios in governing biogeochemical processes, particularly the C:N:P ratio in suspended particulate organic matter (POM), with somewhat less attention given to exported POM and dissolved organic matter (DOM). Herein, we extend the discussion on ecosystem C:N:P stoichiometry but also examine temporal variation of stoichiometric relationships. We have analysed elemental stoichiometry in the suspended POM and total (POM + DOM) organic matter (TOM) pools in the upper 100 m, and in the exported POM and sub-euphotic zone (100 - 500 m) inorganic nutrient pools from the monthly data collected at the Bermuda Atlantic Time-series Study (BATS) site located in the western part of the North Atlantic Ocean. C:N and N:P ratios in the TOM were at least twice that in the POM, while C:P ratios were up to five times higher in the TOM compared to that in the POM. Observed C:N ratios in suspended POM were approximately equal to the canonical Redfield Ratio (C:N:P = 106:16:1), while N:P and C:P ratios in the same pool were more than twice the Redfield Ratio. Average N:P ratios in the subsurface inorganic nutrient pool were ~26:1, squarely between the suspended POM ratio and the Redfield Ratio. We have further linked variation in elemental stoichiometry with that of phytoplankton cell abundance observed at the BATS site. Findings from this study suggest that elemental ratios vary with depth in the euphotic zone mainly due to different growth rates of cyanobacterial cells. We have also

examined role of the Arctic Oscillation on temporal patterns in C:N:P stoichiometry. This study strengthens our understanding of the variability of elemental stoichiometry in different organic matter pools and should improve biogeochemical models by constraining the range of non-Redfield stoichiometry and the net relative flow of elements between pools.

Keywords: North Atlantic Ocean, BATS, Biogeochemistry, Phytoplankton, Stoichiometry

1. Introduction

Nitrogen (N) and phosphorus (P) are critical elements that control primary production in large portions of the surface ocean. Traditionally, N is considered a proximate and P is an ultimate limiting nutrient in surface waters (Tyrrell, 1999), but primary production in the North Atlantic Ocean has been suggested to be P stressed (Wu et al., 2000; Karl et al., 2001; Sañudo-Wilhelmy et al., 2001; Lomas et al., 2010). Alfred C. Redfield first noted the similarity between N:P ratios in surface ocean particulate organic matter (POM) and in deep-water inorganic nutrients; this observation was further extended to include carbon (Redfield, 1934). Oceanographic studies have consistently found mean plankton biomass to adhere to the Redfield Ratio (C:N:P = 106:16:1; Redfield, 1958; Copin-Montegut and Copin-Montegut, 1983; Geider and La Roche, 2002), and since then this ratio has become a fundamental tenet in marine biogeochemistry. Deviations from the canonical ratio have been used to provide insights into phytoplankton physiology (Goldman et al., 1979; Quigg et al., 2003), nutrient limitation of primary production (e.g., Falkowski and Raven, 1997; Moore et al., 2013), efficiency of biological carbon sequestration in the ocean (Sigman and Boyle, 2000) and the input/output balance of the marine N cycle (e.g., Gruber and Sarmiento, 1997). Geochemists use the Redfield conceptual model to determine the state of the marine N cycle using the N* proxy (e.g., Gruber and Sarmiento, 1997). In the context of this proxy, subsurface nutrient N:P ratios > 16:1 suggest net nitrogen gain, while ratios < 16:1 suggest net nitrogen loss (e.g., Gruber and Deutsch, 2014). However, this relatively simple point of view has been shown to yield up to four-fold overestimation of N_2 fixation rates when compared to directly measured rates (Mills and Arrigo, 2010). In part, this overestimation is due to the production and sedimentation of non-N_2 fixer biomass that can occur at ratios much greater than Redfield, particularly in the subtropical and

tropical oceans (Singh et al., 2013; Martiny et al., 2013; Teng et al., 2014). Furthermore, an ocean circulation model has shown that the N:P ratio of biological nutrient removal varies geographically, from 12:1 in the polar ocean to 20:1 in the sub–Antarctic zone, regions where N_2 fixation is not thought to be important (Weber and Deutsch, 2010). With a better understanding of N cycle processes, the validity of the Redfield model for nutrient uptake has been questioned (Sañudo-Wilhelmy et al., 2004; Mills and Arrigo, 2010; Zamora et al., 2010).

Biologically speaking, a fixed N:P ratio, like the Redfield Ratio, would suggest that nutrients are taken up in that ratio during production of new organic matter (Redfield, 1958; Lenton and Watson, 2000). This conceptual model has been challenged by the fact that the variability in nutrient requirements is related to the functioning and evolution of microbes (Arrigo, 2005). The N:P ratio in phytoplankton need not be in the canonical ratio and can vary widely from coastal upwelling to transitional to oligotrophic regions of the ocean. The observed ratio varies with taxa and growth conditions (Arrigo et al., 1999; Quigg et al., 2003; Klausmeier et al., 2004). For example, it has been shown that non–Redfield nutrient utilization is common during blooms (Arrigo et al., 1999) and in regions dominated by cyanobacteria (Martiny et al., 2013). The N:P ratio of *Synechococcus* and *Prochlorococcus*, small and abundant phytoplankton cells in the open ocean, varies from 13.3 to 33.2 and 15.9 to 24.4, respectively, during exponential growth, while the ratio can be as high as 100 during PO_4^{3-} limited growth (Bertilsson et al., 2003; (Heldal et al., 2003). Another cyanobacteria, the N_2 fixer *Trichodesmium* has an N:P ratio that varies from 42 to 125 (Karl et al., 1992), while in general diatoms have a ratio of ~11:1 (Quigg et al., 2003; Letelier and Karl, 1996; Mahaffey et al., 2005). Excess downward dissolved organic nitrogen (DON) fluxes relative to NO_3^- are associated with *Trichodesmium*

abundance (Vidal et al., 1999). Thus the relative abundance of different phytoplankton functional groups may lead to coupling of N and P cycles in non-Redfieldian proportions.

Considerable effort has been made to understand the variability and controls on the N:P ratio in the dissolved inorganic nutrient pool (e.g., Gruber and Sarmiento, 1997; Pahlow and Riebesell, 2000; Arrigo, 2005). In contrast, analysis of C:N:P ratios in particulate organic matter (POM) and dissolved organic matter (DOM) are more scarce (Karl et al., 2001; Letscher et al., 2013). The C:N:P ratio however, has great relevance in oceanography, as it connects the 'currency' of the ocean, i.e., carbon, to some of its controlling variables, N and P. Here, we present a detailed analysis of C:N:P stoichiometry of POM and TOM along with N:P stoichiometry of dissolved inorganic nutrients at the Bermuda Atlantic Time-series Study (BATS) for an eight year period. The observed ratios are correlated with and discussed in the context of co-measured biological parameters such as cell abundances of different phytoplankton groups and chlorophyll *a*. The goal of this study was to quantitatively assess C:N:P ratios in all (POM, TOM and inorganic nutrients) the pools and their deviations from the Redfield Ratio, and relationships to biogeochemical cycling.

2. Methods

2.1 Data Availability

Comment [MWL1]: New subheadings to break the methods into logical units.

Since 1988, the BATS site, located in the western subtropical North Atlantic Ocean (31° 40'N, 64° 10'W), has provided a relatively unique time–series record of nutrient biogeochemical cycles. However, data on total organic C (TOC), total organic N (TON) and total organic P (TOP) and particulate organic C (POC), particulate organic N (PON), and particulate organic P (POP) have only been collected concurrently since 2004. These data were collected from seven

130 different depths (5, 10, 20, 40, 60, 80 and 100 m) over the euphotic zone. We obtained these data
131 from the BATS website (bats.bios.edu) and analysed the data record from 2004-2012.
132
133 **2.2 Analytical Methods**
134 Samples for nitrate (NO_3^-) and phosphate (PO_4^{3-}) were gravity filtered (0.8 μm) and
135 frozen (-20°C) in HDPE bottles until analysis (Dore et al., 1996). NO_3^- and PO_4^{3-} were measured
136 using a Technicon autoanalyser with an estimated inaccuracy of ~0.12 μmol kg^{-1} and 0.02 μmol
137 kg^{-1}, respectively (Bates and Hansell, 2004). The Magnesium Induced Co-precipitation
138 (MAGIC) soluble reactive P (SRP) method (Karl and Tein, 1997) was used starting in late 2004
139 to improve both the sensitivity and the accuracy of the inorganic PO_4^{3-} analysis (Lomas et al.,
140 2010). POC and PON samples were filtered on pre-combusted (450° C, 4h) Whatman GF/F
141 filters (nominal pore size 0.7 μm) and frozen (-20°C) until analysis on a Control Equipment 240-
142 XA or 440-XA elemental analyzer (Steinberg et al., 2001; Lomas et al., 2013). POP was
143 analyzed using the ash-hydrolysis method with oxidation efficiency and standard recovery
144 checks (Lomas et al., 2010). TOC and TON concentrations were determined using high
145 temperature combustion techniques (Carlson et al., 2010). Total P (TP) concentrations were
146 quantified using a high temperature/persulfate oxidation technique and TOP calculated by
147 subtraction of the MAGIC-SRP value (Lomas et al., 2010). Ideally DOM concentrations would
148 have been estimated by subtracting POM from its total organic concentrations, e.g., [DOC] =
149 [TOC] - [POC], but we did not have paired TOC (and TON) and POC (and PON) values;
150 corresponding POC (and PON) values were taken at slightly different depths but on the same
151 sampling day. Nevertheless, subtraction would not have had a substantial impact because, on
152 average, POC and PON values in the upper 100 m were <4% of TOC and TON, respectively

153 (Fig. 1). Both the accuracy and precision of dissolved organic compound concentrations decrease
154 with depth as concentrations of inorganic nutrients increase to dominate the total pools.
155 Chlorophyll *a* pigments were analyzed by HPLC using the method of van Heukelem and
156 Thomas (2001). Samples for flow cytometric enumeration of pico- and nano-plankton were
157 collected on each cruise and analysed as described in Lomas et al. (2013). Export fluxes of POC,
158 PON and POP were estimated using surface-tethered particle interceptor traps deployed at 200 m
159 depth as described in previous publications (Lomas et al., 2010; Steinberg et al., 2001).
160 Elemental masses of material captured in sediment traps, trap collection surface area and
161 deployment length were used to calculate fluxes (see Lomas et al., 2013 for a more detailed
162 methodology on all the described parameters in the method section).
163
164 **2.3 Data Processing**
165 Our POM and TOM analysis was restricted to the upper 100 m, which also reflects the
166 approximate mean depth of the euphotic zone at BATS (Siegel et al., 2001) and the zone where
167 nutrients are depleted to near analytical detection. All data presented as elemental ratios are in
168 mol/mol units. Mixed layer depth was defined as a 0.125 kg m^{-3} difference in seawater density
169 from the surface (Gardner et al., 1995). While mixed layer depths (MLD) were always deepest
170 during winter, the exact timing of the deepest mixing shifted between years. For example, during
171 2005, the MLD was deepest in March, while it was deepest during February in 2006. Therefore,
172 when presenting data on an annual cycle, we aligned our data to the measured timing of deep
173 mixing in each year and combined all the data to a single 12 month composite (e.g., Carlson et
174 al., 2009). Generally the mixed layer depth was no deeper than ~25m in summer, thus we used

this depth range, 0-25m, to represent the 'surface' data and present our analysis in two depth bin, 0-25m and 25-100m.

3. Results

We present time-series data of chemical constitutes in POM and TOM pools (Fig. 1). We further calculated depth-averaged ratios of the chemical constitutes. We first calculated average concentration of each element over the depth segment (e.g., 0-25 m) and then calculated the ratios based upon those averages. Over the entire length of the time-series, euphotic zone TON:TOP ratios varied between 34 and 130 (Fig. 2a), while TOC:TOP ratios varied between 450 and 1952 (Fig. 2b), and TOC:TON varied between 11 and 17 (Fig. 2c).

Suspended euphotic zone PON:POP ratios were generally lower than TON:TOP ratios (Fig. 2, Table 1). The PON:POP ratio ranged from 7 to 140. Similarly POC:POP ratios were much lower than TOC:TOP, varying from 45 to 532. The POC:PON ratio ranged between 1 and 19. Elemental ratios in the TOM and POM were significantly greater than the Redfield Ratio ($p < 0.05$; z test) with the exception of the POC:PON ratio.

3.1. Annual patterns

3.1.1 Concentrations of POM and TOM

There were annual oscillations in POM pools in the upper 100 m (Fig. 1). TOC also showed annual oscillations, however, TON concentrations were relatively constant throughout the study period. The pattern of TOP was an increasing trend during early 2007 until early 2008 (TOP = 0.0936 × decimal year - 187.8; $r^2 = 0.77$, $p < 0.05$). However, there were no long term sustained changes in concentration of POM and TOM.

198

199 **3.1.2 C:N:P ratios in POM and TOM**

200 There were no discernible year-over-year trends in the POM stoichiometry (Fig. 2).
201 Amplitude of variation in the C:N:P ratios of POM was less than that in TOM. TON:TOP and
202 TOC:TOP ratios showed a decreasing trend throughout the year 2007 (r^2 = 0.46, p < 0.05),
203 which was due to an increasing trend in TOP concentration in that year (Fig. 1). There was no
204 annual trend in the TOC:TON ratio. Overall, like POM and TOM concentration patterns, there
205 were no long-term sustained changes in TOC:N:P ratios.

206

207 **3.2 Seasonal variations**

208 **3.2.1 Concentrations of POM and TOM**

209 There was greater variability in C and N pools in the 0-25 m range compared to that in
210 the 25-100 m range (Figs. 4 and 5). In the 0-25 m depth range, TOC showed an increasing trend
211 after deep mixing during the following five months before reaching a plateau (~67 µmol kg^{-1}).
212 POC increased in the first month after deep mixing and then decreased during the next two
213 months and remained constant (~2 µmol kg^{-1}) for the rest of the year (Fig. 4a). The pattern in
214 PON was similar to POC, while those in TON and TOC were opposite to each other during the
215 first two months after mixing and then increased until the sixth month (Fig. 4a, b). These higher
216 values of TOC and TON (observed in both 0-25 m and 25-100 m depth segments) in the sixth
217 month might be attributed to the higher occurrence of *Trichodesmium* colonies during August at
218 BATS (Orcutt et al., 2001; Singh et al., 2013). TOP and POP increased during and one month
219 after the deep mixing in the 0-25m depth range (Figs. 4c). Some of these trends (e.g., higher

values of TOC and TON in the sixth month) were also apparent in the 25-100 m depth range but were not as prominent as in the 0-25 m depth range (Figs. 4 and 5).

3.2.2 C:N:P ratios in POM and TOM

TON:TOP (68 ± 9) and PON:POP (36 ± 11) values were greater than the Redfield Ratio ($p < 0.05$) (Table 1). Patterns in TOC:TOP and TON:TOP ratio, and POC:POP and PON:POP were similar to each other (Fig. 6a, b). TOC:TOP (983 ± 168) and POC:POP (210 ± 67) values were much higher than the Redfield Ratio of 106 ($p < 0.05$). TOC:TON (15 ± 0.5) increased for the two months following deep mixing and decreased until the seventh month (Fig. 6c). POC:PON (6 ± 3) increased in the next month after deep mixing, but remained around the Redfield Ratio throughout the year. Minimal variability in concentration and ratios in the 25-100 m depth range suggests confinement of the more dynamic biogeochemical processes to within the mixed layer, i.e. within 0-25 m (Figs. 5 and 7).

3.2.3 N:P ratios in inorganic nutrients

The average NO_3^-:PO_4^{3-} ratio was 25.6 ± 9.1 in the 100-500 m depth range at BATS, which is greater than the Redfield Ratio (Table 1). We excluded data from the top 100 m in this analysis due to low precision relative to the mean nutrient values which are at or near analytical detection limits due to active biological uptake. NO_3^- and PO_4^{3-} were at their highest concentrations before deep mixing and decreased immediately following the month of deepest mixing and remained constant for the rest of the year (Fig. 8). The decrease in NO_3^- and PO_4^{3-} concentrations was likely due to dilution with low nutrient surface water during mixing.

3.2.4 N:P ratios in the particulate flux at 200 m

The PON fluxes increased during and peaked immediately after winter mixing, while POP fluxes showed elevated values before and shortly after the time of deep mixing (Fig. 8). The N:P ratio of export fluxes was nearly twice that of PON:POP ratio in the suspended matter (upper 100 m; Table 1).

3.2.5 Chlorophyll *a* and phytoplankton cell abundance

Chlorophyll *a* values decreased after the spring bloom that was stimulated by deep mixing (Fig. 9a). *Prochlorococcus* was dominant during the oligotrophic period of the year, while these were least abundant around the time of deep mixing (Fig. 9b). In contrast, *Synechococcus* and picoeukaryotes were more abundant during the more productive season (Fig. 9c,d), and followed the annual pattern in Chlorophyll *a*. There was no discernible seasonal pattern in nanoeukaryote abundance (Fig. 9e).

4. Discussion

From the approximately eight years of BATS data presented here, it is apparent that the total and particulate organic matter C:N:P stoichiometries are not a long-term fixed ecosystem property, but vary seasonally and deviate substantially from the canonical Redfield Ratio. Observed C:N:P ratios in TOM and POM were much greater than the Redfield Ratio, averaging 983:68:1 and 210:36:1, respectively, for the entire dataset (Figs. 2, 4, 5).

4.1 Connections among POM, TOM and inorganic nutrients

Comment [MWL2]: Nearly the entire discussion has been reorganized whether within or between sections. I have not highlighted it as the entire discussion ended up highlighted.

Redfield hypothesized what was effectively a two-box model of nutrients shuttling between particulate and dissolved form. However, there are number of different biological, chemical and physical processes acting on particles as they settle through the water column. Higher N:P ratios in the particulate fluxes than in the suspended matter could be due to the preferential export of N or preferential remineralisation of P, but similar C:N ratios in the fluxes and suspended matter would lend more support to the latter scenario (Figs. 4, 8; Table 1; Monteiro and Follows, 2012). The N:P ratio of export fluxes was also generally more than twice that of the dissolved NO_3^-:PO_4^{3-} ratio at depth (Fig. 8c). The preferential remineralization of P from settling material could potentially explain this difference, as there is little evidence for N loss in this well-oxygenated region, however the advective flux of low NO_3^-:PO_4^{3-} waters needs to be considered. Indeed, the literature indicates that sub-euphotic waters at BATS are a mixture of water originated at the north of the site, which has characteristically low NO_3^-:PO_4^{3-} ratios (Bates and Hansell, 2004; Singh et al., 2013). The processes of remineralization are not direct from particulate to inorganic pools and indeed, cycling through the dissolved organic pool, which dominates TOM, is important. One explanation for the TON:TOP ratio being greater than the Redfield Ratio is that TON is less reactive than TOP and broken down mainly in the subsurface layer (Letscher et al., 2013), while TOP is labile or semi-labile and both remineralized and assimilated at a shallower depth (Björkman et al., 2000). Consequently, TOP has faster turnover times (Clark et al., 1998). In contrast to this interpretation, our observations suggest that TON and TOP values increase slightly with depth suggesting a net (i.e., remineralization exceeding assimilation) flow of material from the particulate organic pool to the dissolved organic pool for both elements (comparing data in Figs. 4 and 5).

Our results on the TON:TOP ratio have important implications in ocean biogeochemistry of oligotrophic waters where DON and DOP concentrations in the sunlit layers exceed the concentration of inorganic nutrients by an order of magnitude. Dissolved organic pools are essential in sustaining phytoplankton growth in these regions (Church et al., 2002; Williams and Follows, 1998). Nutrient levels decide phytoplankton growth and their stoichiometry (Klausmeier et al., 2004), hence TON:TOP in the oligotrophic regions might determine optimal N:P stoichiometry of phytoplankton rather than the inorganic pools alone.

4.2 Linkages of concentrations and ratios of POM and TOM to chlorophyll *a* and phytoplankton

We hypothesize that C:N:P ratios in the aggregated phytoplankton community itself changes the elemental stoichiometry of the POM and TOM pools. The C:N:P ratio is different in different phytoplankton communities and their biological uptake and degradation could potentially change the elemental stoichiometry of the particulate and dissolved organic matter. The C:N:P ratio varies geographically and its pattern correlates with global variations in temperature, overall nutrient concentrations and phytoplankton functional groups. These latitudinal patterns in the C:N:P ratio have been attributed to changes in phytoplankton community as polar (colder) regions have a high abundance of diatoms with low N:P and C:P ratios, in contrast to the directly measured high elemental ratios in cyanobacteria from warmer regions (Martiny et al., 2013). So how and why does C:N:P ratio vary in phytoplankton communities? Two mechanisms could explain variability in the C:N:P ratios in a phytoplankton community. The first mechanism suggests that the taxonomic composition of a phytoplankton community influences its elemental composition. Elemental ratios inside a cell are controlled by

310 growth strategies (Klausmeier et al., 2004) . Studies have reported low C:P and N:P ratios in fast
311 growing diatoms (e.g., Price, 2005), whereas slower growing cyanobacteria have C:P and N:P
312 ratios higher than the Redfield Ratio (Bertilsson et al., 2003; Martiny et al., 2013). More
313 precisely, it is not so much the growth rate that determines the difference, but the machinery
314 invested in nutrient acquisition versus protein production.

315 The second mechanism links the nutrient supply ratio to a taxonomically 'hard-wired'
316 cellular elemental ratio (Rhee, 1978). Chlorophyll *a* values were anti-correlated with TOC
317 values ($r^2 = 0.76$, $p < 0.05$). The gradual increase in Chlorophyll *a* during the four months before
318 deep mixing is due to similar increase in MLD before deep mixing (Fig. 3), which suggests that
319 there may be enhanced nutrient flux into the upper layer well before deep mixing (e.g., Fawcett
320 et al. 2014). *Prochlorococcus* and *Synechococcus* profiles were correlated to each other in the
321 first seven months from the point of deepest mixing ($r^2 = 0.58$, $p < 0.05$) and there was no
322 relation in the rest of the year in the 0-25 m depth range. Furthermore, *Synechococcus* cell
323 abundance was correlated with POC ($r^2 = 0.67$ $p < 0.05$), PON ($r^2 = 0.47$ $p < 0.05$), POP ($r^2 =$
324 0.29 $p < 0.05$) and anti-correlated with TOC values ($r^2 = 0.72$ $p < 0.05$) in the 0-25 m depth
325 range. *Synechococcus* is more abundant during the more productive season whereas
326 *Prochlorococcus* is dominant during the highly oligotrophic part of the year. Such patterns are
327 typically observed in many parts of the ocean. The seasonal pattern of picoeukaryote abundance
328 was similar to that of *Synechococcus* ($r^2 = 0.58$ $p < 0.05$) and Chlorophyll *a* ($r^2 = 0.81$ $p < 0.05$).
329 POC:PON:POP ratios in *Prochlorococcus*, *Synechococcus* and picoeukaryote are 234:33:1,
330 181:33:1 and 118:15:1, respectively at the BATS site (Martiny et al., 2013 and Lomas et al.,
331 unpublished data), which clearly suggests imprints of a mixture of *Prochlorococcus*,
332 *Synechococcus* on the observed POM stoichiometry presented in Table 1. Biomass of

333 *Prochlorococcus*, *Synechococcus* and picoeukaryotes together contributes ~40% to the POC pool
334 (Casey et al. 2013) and ~75% to the PON pool (Fawcett et al. 2011), with major contributions
335 from each group varying seasonally. Hence, variability in biological parameters could potentially
336 explain a significant fraction of the variability in the POM and TOM ratios, but not all of it. So
337 what else drives the variability in the C:N:P ratios?

338 We analysed trends in the TON:TOP and TOC:TOP ratios for December 2006 to January
339 2008 data along with phytoplankton cell abundances for the top 100 m BATS data. Since the
340 variation in TON:TOP and TOC:TOP were due to an increasing trend in TOP, we correlated
341 TOP concentrations with a lag of three months (there is a time lag between phytoplankton and
342 elemental abundance as observed by Singh et al., 2013) in phytoplankton cell abundances (data
343 during September 2006 to November 2007; Fig. 10a). We observed significant anti-correlation
344 ($r^2 = 0.61$, $p < 0.001$) between nanoeukaryotes and TOP but the data did not correlate with other
345 phytoplankton groups (Fig. 10a). In the paucity of elemental composition data on
346 nanoeukaryotes, we hypothesize that these cells have a high requirement for P and are potentially
347 meeting that requirement by assimilating TOP.

348 We further analysed this increasing trend in the TOP concentration with climate indices.
349 The Arctic Oscillation is a major climatic phenomenon in the North Atlantic Ocean (Thompson
350 and Wallace, 1999). Positive trends in the Arctic Oscillation lead to higher temperatures,
351 advanced spring, and increased CO_2. This could lead to enhanced uptake of CO_2 during spring as
352 has been found in terrestrial systems (Schaefer et al., 2005). Higher build-up of organic matter
353 would require more P and hence we correlated TOP concentration with monthly Arctic
354 Oscillation index with a lag of a year (monthly Arctic Oscillation indices are from November
355 2005 to December 2006, because there is a lag of one year before climatic oscillations in the

356 North Atlantic show its impact on surface biogeochemistry; Fromentin and Planque, 1996). We
357 observed a significant correlation ($r^2 = 0.46$, $p < 0.01$) between the Arctic Oscillation and TOP
358 concentrations (Fig. 10b). Since variations in phytoplankton cell abundances and climate
359 variability could not explain all the variation in the elemental stoichiometry, other mechanisms
360 are yet to be identified to explain the observed variability in the elemental stoichiometry.
361
362 **4.3 Role of DOM in microbial carbon export**
363 Many biogeochemical model estimates of export production assume Redfield
364 stoichiometry in export fluxes but a non-Redfieldian approach has gained appreciation recently
365 (Letscher and Moore, 2015). Export production is estimated to be 3-4 mol C m^{-2} yr^{-1} in the
366 BATS region (Jenkins, 1982; Emerson, 2014), which requires more nutrient input than
367 observations suggest (Williams and Follows, 1998). A possible mechanism to sustain such
368 export production is the supply of DOM to the sunlit layer.
369 DOM consists of complex compounds whose chemical characterization is incomplete,
370 but it is evident that DOM elemental stoichiometry differs drastically from the Redfield Ratio.
371 Differential production and degradation of DON and DOP with lifetimes comparable to the gyre
372 circulation could potentially change the overall stoichiometry of nutrient supply (Voss and
373 Hietanen, 2013). Preferential degradation of DOP rather than DON expands the niche of
374 diazotrophs beyond that created by subsurface denitrification. Diazotrophs can quickly utilize
375 recycled DOP (Dyhrman et al., 2006). Simultaneously, these diazotrophs release DON
376 (Mulholland, 2007), which can be used by other phytoplankton, but this DON likely has
377 associated DOP. In the P stressed Sargasso Sea, DOP contributes up to 50% of P demand for
378 primary production (Lomas et al. 2010) and up to 70% to the exported POP (Roussenov et al.,

2006; Torres-Valdés et al., 2009). Indeed, a 1-D biogeochemical model for BATS that included an explicit DOP pool and a generic DOM pool significantly improved the capture of natural variability in both particulate (suspended and exported) and dissolved (organic and inorganic) pools (Salihoglu et al. 2007). These model results, as well as others connecting DOP cycling to particulate P export (e.g., Roussenov et al. 2007), suggest a strong need for direct rate measurements of DOM production and assimilation (e.g., Mahaffey et al. 2014).

Conclusion

Our time-series analysis suggests temporal and depth variability in the C:N:P ratio in the Sargasso Sea. C:N:P ratios in the TOM were significantly higher than the canonical Redfield Ratio, while C:N was similar to the Redfield Ratio in the POM. We observed seasonal variability in stoichiometry but on average the TOC:TON:TOP ratio was 983:68:1 and the POC:PON:POP was 210:36:1. Seasonal variation in POM stoichiometry appears to be largely driven by the growth of *Synechococcus* during winter mixing, while flourishing of *Prochlorococcus* cells during the oligotrophic period (fall) could also explain some variability in the stoichiometry. The C:N:P ratio in *Prochlorococcus* cells resembles observed mean POC:PON:POP ratio at BATS (210:36:1). The N:P ratio in subsurface inorganic nutrients was also greater (N:P = 26) than the Redfield Ratio in this region. We observed a significant decreasing trend in TON:TOP and TOC:TOP during 2007, which was due to an increase in TOP concentration and could have been partly driven by the Arctic Oscillation and a decrease in the relative abundance of nanoeukaryotes. Other causes for the observed variations in the elemental stoichiometry need to be explored; however, this elemental stoichiometry analysis may improve biogeochemical models, which have hitherto assumed Redfield stoichiometry to estimate export fluxes.

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403

404

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Comment [MWL3]: A number of relevant references added in conjunction with the rewrite of the discussion.

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603 **Table 1.** Average concentration (μmol kg^{-1}), molar ratio of various biogeochemical parameters

604 and particle fluxes (mmol m^2 d^{-1}) from the BATS data presented in Fig. 1.

Concentration (C) in the upper 100 m			
Parameter	C ± σ*	no of samples	Sampling period
TOC	63.81 ± 2.86	714	Jan 2004 - Dec 2011
TON	4.43 ± 0.50	712	Jan 2004 - Dec 2011
TOP	0.07 ± 0.03	547	Jun 2004 - Nov 2011
POC	2.36 ± 1.14	844	Jan 2004 - April 2012
PON	0.40 ± 0.19	845	Jan 2004 - April 2012
POP	0.01 ± 0.01	696	Jan 2004 - April 2012
Ratio (R)† in the upper 100 m			
Parameter	R ± σ	no of data points€	Sampling period
TOC:TON	15 ± 0.5	86	Jul 2004 - Dec 2011
POC:PON	6 ± 3	95	Jan 2004 - Apr 2012
TON:TOP	68 ± 9	77	Jul 2004 - Nov 2011
PON:POP	36 ± 11	88	Jan 2004 - Apr 2012
TOC:TOP	983 ± 168	78	Jul 2004 - Nov 2011
POC:POP	210 ± 67	88	Jan 2004 - Apr 2012
Inorganic nutrient stoichiometry in 100-500 m			
Parameter	(C or R) ± σ	no of data points	Sampling period
NO_3^-	2.74 ± 2.40	3425	Oct 1988 - July 2012
PO_4^{3-}	0.11 ± 0.13	3405	Oct 1988 - July 2012
NO_3^-:PO_4^{3-}	25.6 ± 9.1	2415	Oct 1988 - July 2012
Particle fluxes at 200 m			
Parameter	C ± σ	no of samples	Sampling period
C	1.68 ± 1.07	254	Jan 1989 - Dec 2011
N	0.23 ± 0.16	254	Jan 1989 - Dec 2011
P	0.008 ± 0.014	64	Oct 2005 - Dec 2011
Ratio in particle fluxes at 200 m			
Parameter	R ± σ	no of data points	Sampling period
N:P	57 ± 46	61	Oct 2005 - Dec 2011
C:P	287 ± 269	62	Oct 2005 - Dec 2011
C:N	7.9 ± 2.8	252	Jan 1989 - Dec 2011

605 *σ is standard deviation of the samples mentioned in next the column. †Ratios and their standard
606 deviations are derived from the monthly mean values (€one datum would be mean of many
607 values of concentration for a particular month) of concentration in the upper 100 m.
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Figure Captions:

Fig. 1. Monthly BATS data on C, N and P in total and particulate organic matter in top 100 m during Jan 2004 to April 2012.

Fig. 2. Monthly stoichiometry during 2004-2010 at 0-100 m. Solid lines are three month running means. Error bars are 1σ standard deviations from the mean values.

Fig. 3. Mixed layer depth (MLD) during the sampling period at BATS site.

Fig. 4. Box/whisker plot comparing the annual concentrations of total (open bars) and particulate organic matter (filled bars) relative to the deep mixing in 0-25 m depth at BATS (data used from January 2005 - December 2011). Bottom and top of the box define the 25% and 75% data distribution, and the 'error' bars define the 5% and 95% data distribution. The dark gray vertical bar represents the period of deep mixing (DM) for each year.

Fig. 5. Box/whisker plot comparing the annual concentrations of total (open bars) and particulate (filled bars) matter relative to the deep mixing at 25-100 m depth (data used from January 2005 - December 2011). All else as in Figure 4.

Fig. 6. Box/whisker plot comparing the annual ratios of elemental stoichiometry relative to the deep mixing at 0-25 m depth (data used from January 2005 - December 2011). All else as in Figure 4.

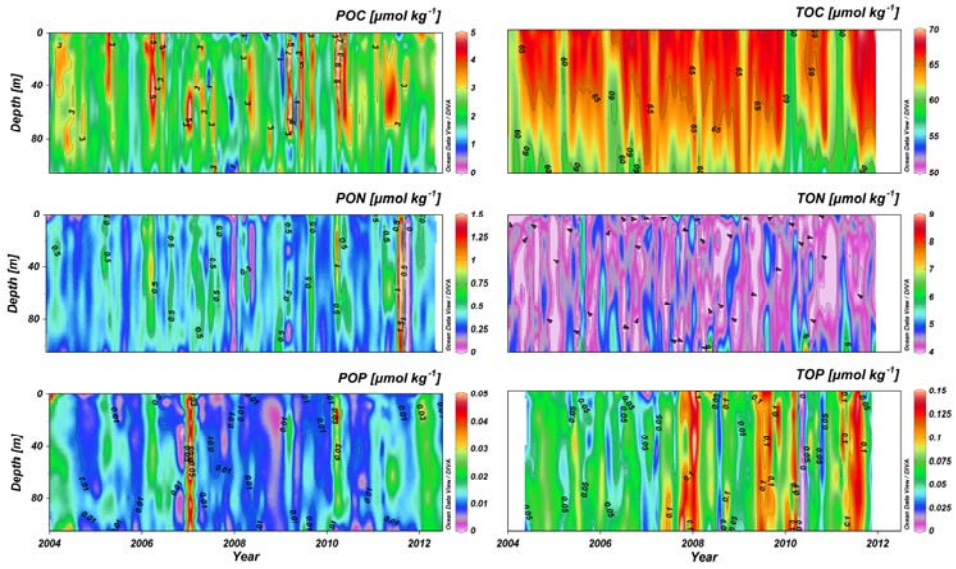
Fig. 7. Box/whisker plot comparing the annual ratios of elemental stoichiometry relative to the deep mixing at 25-100 m depth (data used from January 2005 - December 2011). The gray bar represents the period of deep mixing (DM) for each year. All else as in Figure 4.

Fig. 8. Box/whisker plot comparing the annual variation of NO_3^- and PO_4^{3-} and their ratio relative to the deep mixing at 100-500 m depth (data used from January 2005 - December 2011). The gray bar represents the period of deep mixing (DM) for each year. All else as in Figure 4.

Fig. 9. Box/whisker plot comparing the annual variation in Chlorophyll *a* and cell counts for *Prochlorococcus*, *Synechococcus*, Picoeukaryotes, and Nanoeukaryotes relative to the deep mixing in 0-25 m depth at BATS (data used from January 2005 - December 2011). The gray bar represents the period of deep mixing for each year. All else as in Figure 4.

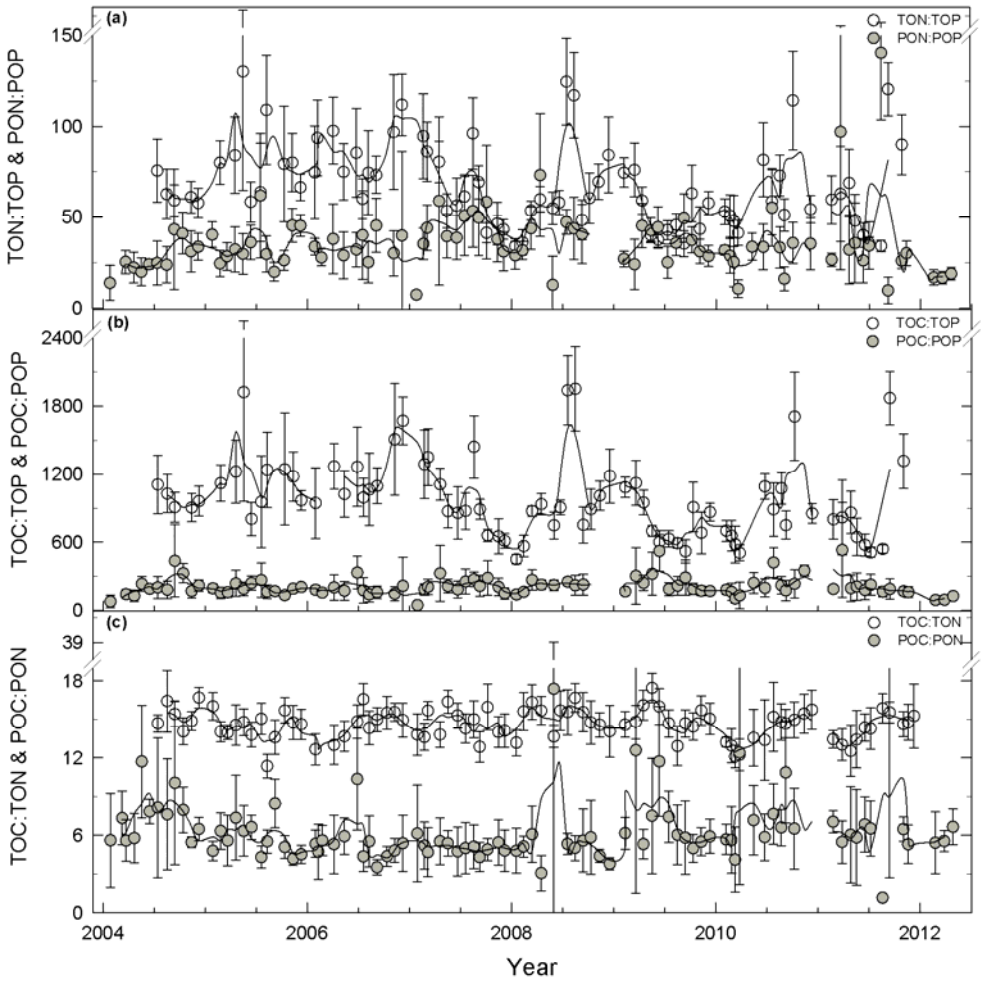
Fig. 10. Relationship between TOP (Dec 2006 - Jan 2008) and (a) cell abundances (natural log transformed) of *Prochlorococcus*, *Synechococcus,* Picoeukaryotes and Nanoeukaryotes during Sep 2006 - Nov 2007. Among cell abundances, only Nanoeukaryotes showed a significant relationship with TOP (r^2 = 0.61, p < 0.001) (b) Relationship between TOP and Arctic Oscillation index during Nov 2005 - Dec 2006 (r^2 = 0.46, p < 0.01).

Fig.1



Comment [MWL4]: Color figure to improve value.

673 **Fig. 2**



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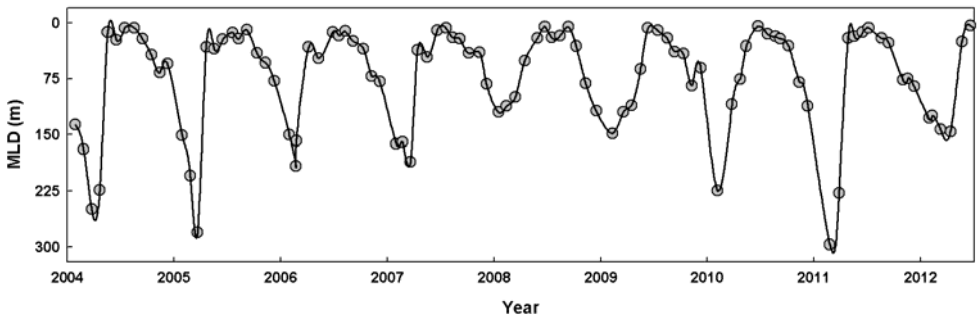
680 **Fig. 3**



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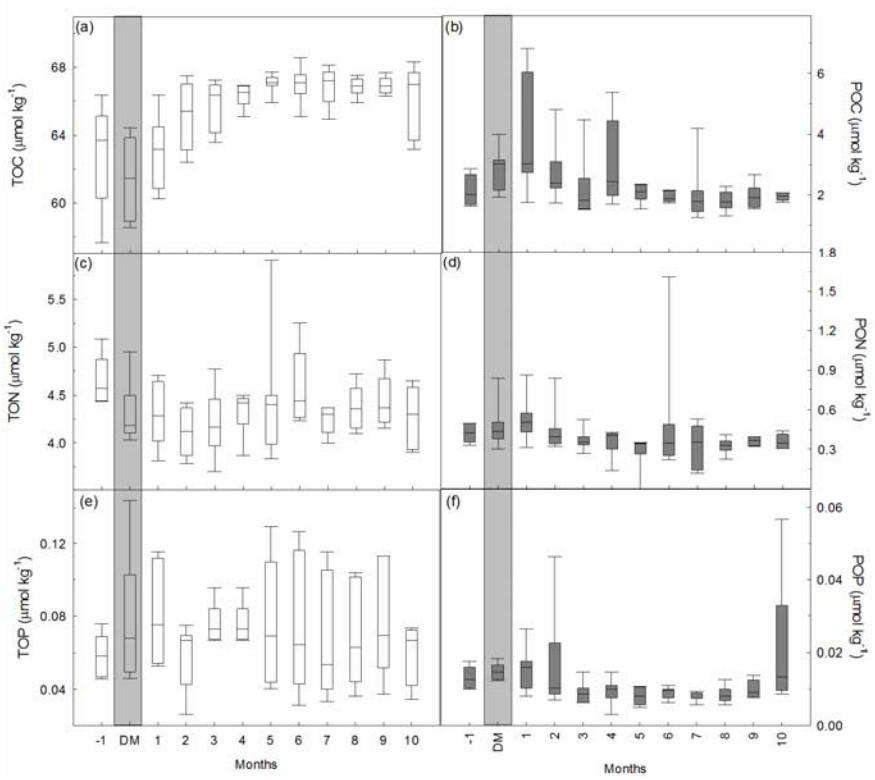
698 **Fig. 4**

Comment [MWL5]: New layout for figures 4-8 to improve clarity of the figure.



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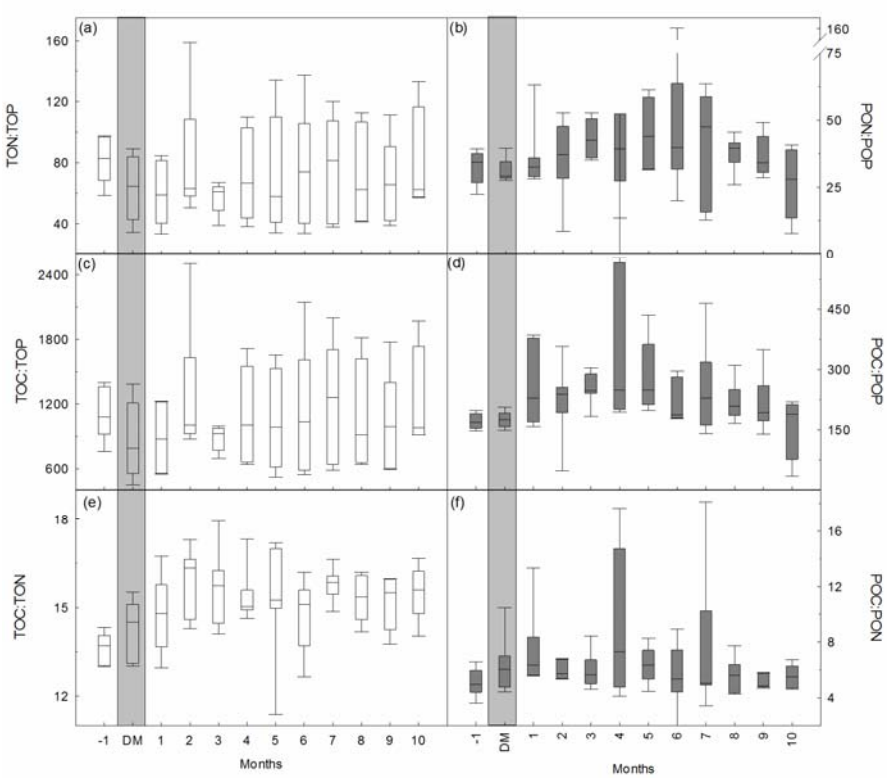
704 **Fig. 5**



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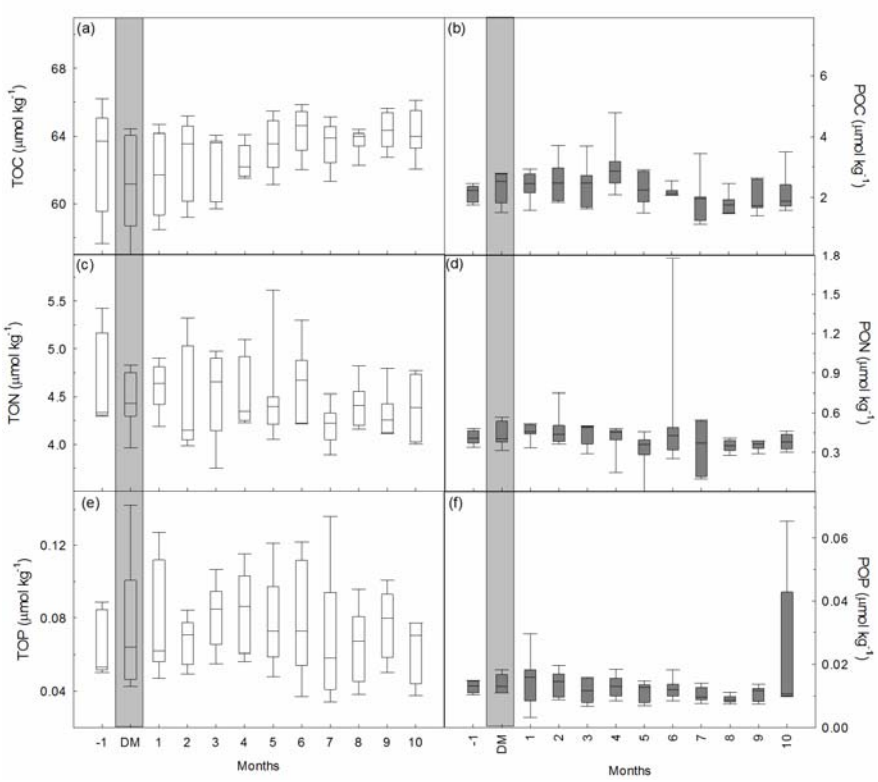
710 **Fig. 6**



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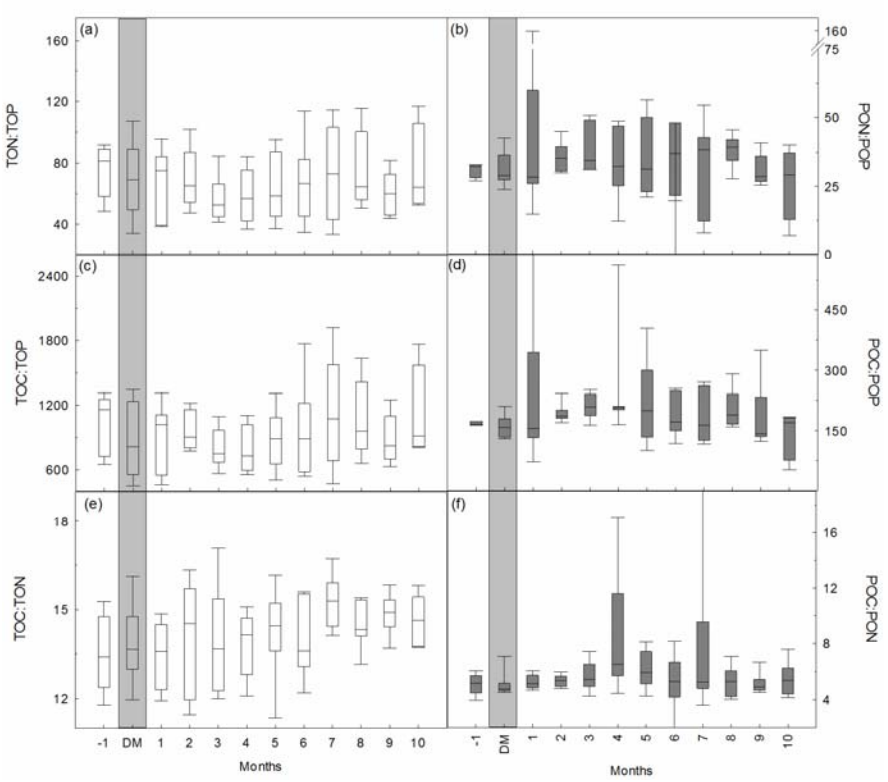
716 **Fig. 7**



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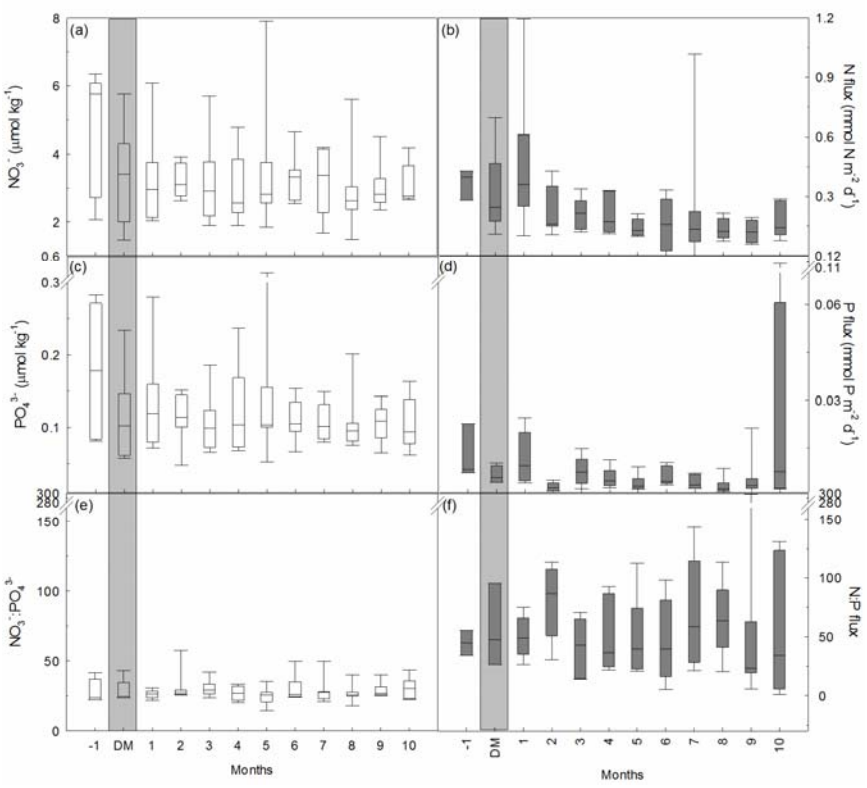
722 **Fig. 8**



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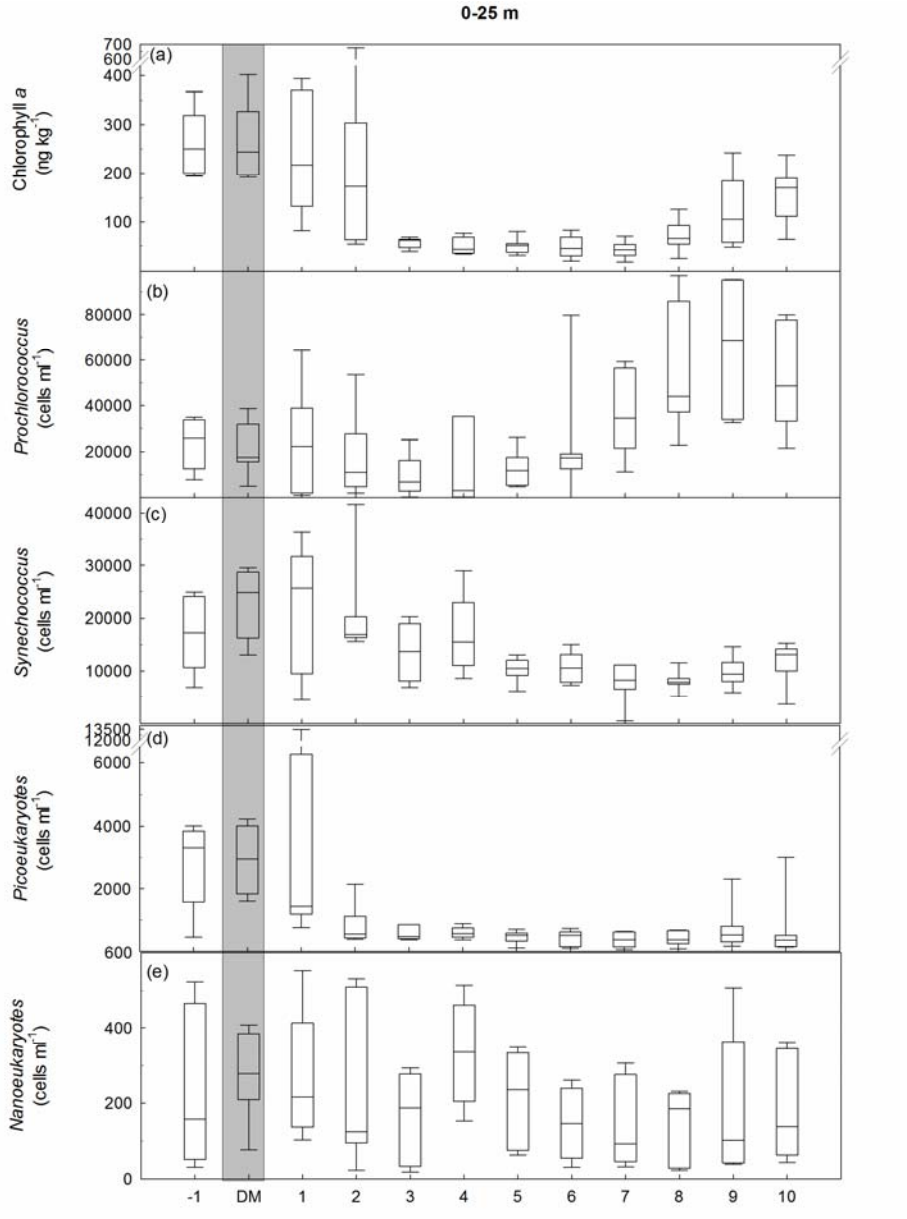
727 **Fig. 9**



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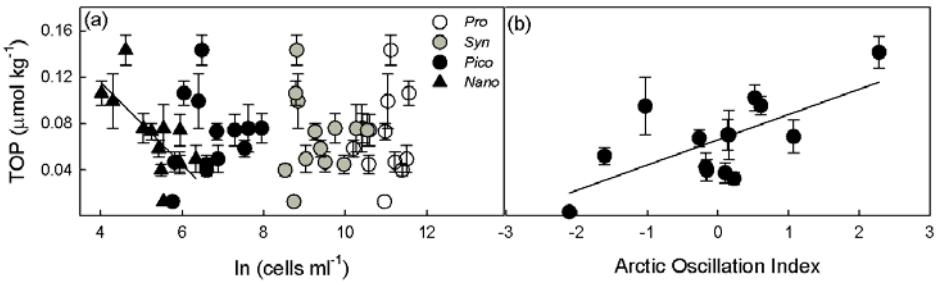
730 **Fig. 10**



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