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.