<center>**Point-by-Point Responses and Accompanying Revisions to BG-2016-511**</center>

We thank both reviewers for their useful comments. We have considered all of their comments and incorporated a number of changes into the manuscript as part of these minor revisions, which has improved it considerably. We provide point-by-point responses below.

Reviewer #1: All suggested comments from Dr. Shelley were incorporated, we thank her for her careful reading and suggestions. Because the upper water column has such a small inventory of dissolved cobalt, we felt a brief mention regarding the massive increase in use of lithium-cobalt batteries and cobalt mining near the South Atlantic region would be particularly relevant to this manuscript, and we have added two sentences to the introduction based on this, connecting the potential impact the small inertia of the upper water column cobalt inventory.

Reviewer #2: We thank Dr. Boye for her comments on the manuscript. We provide point-by-point responses here, although we were not able to obtain a digital (non-pdf) version of the review so reviewer comments are paraphrased or quoted. In general, we have incorporated the reviewer's suggestions or in some cases there was a misunderstanding about the data or interpretation they are interested in being elsewhere in the paper. I think the major scientific point of this paper is that dissolved Co : phosphate ratios are extraordinarily diverse, far more so than for any other macro or micronutrient by a large margin. This paper attempts to capture the full extent of this stoichiometric diversity particularly in its effect on the dissolved phase, with novel connections to the particulate and biochemical phases. To do so required employing some new approaches using simple statistical methods, and using the power and confidence associated with a large number of statistical analyses rather than stringent methods on single or few analyses of a large dataset as done previously. This focus was highlighted further by some additional sentences in the introduction and altered sentences in the abstract.

The reviewers ask about the methods used: "all this approach based on multifactorial calculations of the processes is indeed little convincing without the detail and the methodology of the used calculations". There are three minor issues here. First, regarding the methods, there is no multifactorial analysis, simply a Matlab script that repeats a simple two-way linear regression many times on groupings of 5 depths many times across the sections in order to achieve the high-resolution stoichiometry targeted in this study. This method is simple and is fully documented in the methods. It could be conducted in Excel or similar software for each 5 data-point analysis, the script serves to iterate the analysis many times over.

Second, the reviewer asked for additional information about the length of the vectors. The length of vectors shown in Figure 3 were purposefully made to be larger than the plot since they do not connote information in this context, and this is stated now in the caption. The vector addition used in Figure 10 is documented in the paper already in the discussion, and is used primarily as an example scenario (e.g. what vectors could be added to result in the observed negative vector). We agree with the reviewer that this could become a useful quantitative analysis in the future, but a detailed characterization of vector length and their meanings is likely beyond the scope of this manuscript.

Third, regarding the question about the linear regression approach, the reviewer suggests increasing the threshold from r= |0.7| to |0.88| and the number of data points per regression from 5 to 8. In response, we point out that this exact broader scale analysis with more data and higher r2 values was conducted

and presented in this manuscript as described in Section 3.2 as "aggregate" datasets (and the regressions figures themselves are shown in Noble et al., 2017 BG, their Figure 11, r2 varying from 0.67 to 0.92). After presenting these large scale and more significant features, we argue that there is a large amount of upper euphotic zone data that was excluded in order to acquire these high r2 regressions, yet likely represents real elevated stoichiometric features. This "profile-based" analysis approach was applied to measure these shallow phenomena that we have long observed, but that simple aggregate linear regressions have been unable to capture effectively. We point out that the reviewer's recent paper also observed the same phenomenon, where in the Dulaquais et al., 2014 dataset for the North Atlantic Gyre, their *suggested value of r2 of greater than 0.77 would exclude their own Co:P value for this region* (their figure 8, R2= 0.65 for n=32). Visually the acceleration of Co:P stoichiometries in the upper photic zone is also apparent in their dataset as well (their Figure 8 again). As a result, we agree with their suggestion that this finer approach to characterizing stoichiometries could be useful on multiple section analyses (but decline to include more data in this already large manuscript).

This demonstrates the motivation for conducting the moving depth window regression approach in our study, in that trying to fit a single line to a stoichiometry that is accelerating creates errors and hence we argue that our combination of aggregate dataset and moving window regressions is able to capture the bulk and fine-scale features within the dynamic Co:P ratios in the upper water column. The high resolution approach requires smaller depth intervals in order to capture the resolution because they are increasing (and accelerating) towards the surface. For many years we have observed a significant increase in slope in the upper photic zone which we have believed to be a real biochemical effect. With sectional datasets now available, we are able to combine dissolved, particulate, and biochemical protein datasets in this manuscript to study this phenomenon. For the dissolved samples to be expanded to 8 depths from 5 would result in a loss of depth resolution in which these features would be obscured (this effectively imposes an assumption of a constant stoichiometry rather than an increasing one). We have altered our Figure 3 to present all of the r value data that is above our threshold to be fully transparent to the reader. To change to 8 points and arbitrarily higher r values thresholds is ignoring the motivation for this study to tease a subtle but apparently widespread signal from the euphotic zone, which is also supported by particulate and protein data here and which is found across two ocean sections.

The reviewer also comments on how: "Only the middle point is used in vector analysis". This is a misunderstanding. For each 5-depth point regression the resulting slopes are plotted centered around the data, which is the midpoint within the 5-points. We could easily switch the choice of depth to be any of the 5-depth points within the regression, but centering at its midpoint seemed most logical.

The reviewer's second main concern is regarding the lack of consideration of a mixing vector in the vector analysis. The reviewers incorrectly state that we "argue that only remineralization and scavenging govern the deep distribution of cobalt in the North Atlantic". On the contrary we state that "These scavenging signals co-occur with distinct water masses identified by OMPA analysis, implying that these scavenging processes are being integrated on decadal-to-century timescales of deepwater circulation processes within the ocean interior." While it is true that water mass mixing could alter the ratio of Co and P, mixing itself is not a process that removes or add Co or P from the dissolved phase (except in estuaries). Moreover, the ratios of the dCo and PO4 provided in the review produce an almost identical ratio of 32 and 31-35 (40, 70-80pM dCo; 1.25 and 2.25 PO4), so mixing these NADW and AABW water masses would not create a significant vector in Co:P space by their example. Our point here is that in these localized fine scale regression analyses vertical processes of uptake remineralization and

scavenging, whose signals are clearly integrated through horizontal advection, can explain the large variation and change in sign of the Co:P slopes. Finally, it is not clear that mixing would produce a single vector depending on the ratios being mixed, but instead could be in a number of directions. We did include a mixing vector in our original vector diagram for upwelling (Noble et al., 2008), but that may be a specific coastal scenario. As a result, we politely decline to include a mixing vector in the schematics at this time and instead focus on the chemical and biochemical processes that are capable of moving dissolved cobalt and phosphorus to the solid phase, rather than mixing processes that cannot.

The reviewer suggests using the water mass analysis in this study of end member distributions: "since the complete analysis of water-masses is available, as performed by Jenkins et al., 2015), the combination of end-members weighted by the proportion of each water-mass will help to determine if a located depth is a source (remineralization) or a sink (scavenging) for dCo in the deep sea." We point out that this water analysis has already been included and is shown in Figure 2 for the North and South Atlantic Zonal sections including the following text that relates to the reviewer's suggestion. Our manuscript states: "These scavenging signals co-occur with distinct water masses identified by OMPA analysis, implying that these scavenging processes are being integrated on decadal-to-century timescales of deepwater circulation processes within the ocean interior (Noble et al., submitted). Specifically, negative slopes water masses were found to be in the Denmark Straits Overflow Water/Antarctic Bottom Water/Iceland Scotland Overflow Water (DSOW/AABW/ISOW) and Classical Labrador Seawater (CLSW; Fig. 2) water masses both of which have long deepwater transit times (Jenkins et al., 2015)." We argue that the negative vectors we observe are the influence of slow scavenging processes accumulating on isopyncals during advection, and hence that the influence of scavenging and mixing are combined in the negative vectors we observe. The reviewers imply that mixing of Co is a conservative process (without any scavenging), but we respectively disagree with this argument from Dulaquais et al., 2014, and argue that the data shown here, and in a more substantive manuscript on Co scavenging (Hawco et al, Marine Chemistry in revision) provide evidence of mesopelagic scavenging. In particular, in the latter manuscript a comparison of dCo with C14 ages in DIC showed basin scale scavenging processes between the Atlantic and Pacific basins. Much of the reviewer's interest in scavenging is more pertinent to the Hawco et al. manuscript, while the present manuscript focuses on the dynamic variation in dCo:P relationships in the vertical dimension, which must include some discussion on the unusual negative slopes in the mesopelagic and deep ocean and how the shift from remineralization to scavenging can result in the transition in positive to negative slopes as shown in Figure 10.

The reviewer states that we are inconsistent with Moffett and Ho's Co and Mn microbial oxidation dataset which did not find measurable Mn oxidation at BATS. This is incorrect, Moffett and Ho found phytoplankton uptake dominates within the euphotic zone at 60m, which is consistent with our uptake/remineralization vector based on positive slopes at this depth. Moffett and Ho did not present uptake rates in the mesopelagic in that manuscript.

The reviewer suggests an alternate means for particulate Co-Mn correlations, "The strong Co-Mn correlation can be interpreted in another way than that actually proposed: it is indeed possible that the biogenic Co fraction is remineralized while the authigenic fraction (scavenged fraction) is not (i.e.; the one way trip of Co presented by authors)". We appreciate the reviewers considering and describing this scenario in detail. This interpretation is actually the phenomenon we are presenting and arguing for in every aspect they discuss (an increasing importance of scavenged/authigenic particulate for pCo and

pMn with depth resulting in the pCo:pMn relationship and the increased pCo:pP relationship). We are glad to hear they agree! We point out that it is this authigenic pCo formation process that in turn creates the negative dCo:dPO4 slopes in the dissolved phase we have been debating above. We have found that the connections between dissolved and particulate phases are fascinating yet can also be confusing and take time to fully consider given the mirroring/opposite effects the phases can have on each other and out community's limited experience in direct comparisons of dissolved and particulate data.

The reviewer states that the vector approach does not take into account differential remineralization. This is an interesting point, but upon discussion we think it is not true. The profile-based regression analysis does not include any a priori assumption about stoichiometry or remineralization efficiency. It is simply a measurement of dCo and dP change in response to each other in the vertical dimension. There were no Co* values reported in Hawco 2016 or Noble 2012, but it is here. I understand the hypothesis: an alternate explanation for vertical stoichiometric structure observed here is the preferential remineralization of P over Co creating the Co deficiency, as an alternative to scavenging. This is a useful alternate perspective that we have added to the discussion. Reflecting on it, there are several datasets that argue against differential remineralization being the major driver in dCo distributions: 1) the scavenged like profile is inconsistent with this (implying removal rather than slowed accumulation), 2) the correlation of pCo and pMn in the mesopelagic is consistent with biotic Mn oxidation rather than with slow Co remineralization (which would not create a correlation with Mn), and 3) the negative slopes of dCo:P are consistent with Co loss rather than slowed accumulation (which would be a positive slope still). To clarify, the prior and current observation of increased dCo in the OMZ plumes were not interpreted as a result of slowed / differential remineralization but reduced Mn and Co scavenging. This is an area that is ideal for future investigations that are ongoing by collaborators.

The reviewer suggests incorporating data from Dulaquais et al into the statistical analysis of this study and the table for aggregate analysis. We have incorporated the reviewer's published Co:P ratios into Table 1, and we thank her for this suggestion. As for incorporating other sections of data into the vector and profile-based regression analysis, that is beyond the scope of what we can conduct in this study. There is already a lot of data within this study and there were some intercalibration challenges for dCo at the western portion of the North Atlantic, as the reviewer is aware. Future efforts could apply this approach to other ocean sections of Co:P.

The reviewer comments on our choice of a Co* ratio from the pCo:pP North Atlantic ratio, and argues that it should instead use the lower Twining from phytoplankton species. This is a good comment and one we also wrestled with. We point out that Redfield et al's original studies used oceanic particulate carbon, nitrogen and phosphate ratios as well, and they have been interpreted as being reflective of the aggregate community, so there is precedent for working from the aggregate particulate phase. The cell specific numbers from Twining et al. are taxon-specific and span 140-3000 depending on the group (their Table 5). We were transparent about the challenge of selecting a single Co:P in the manuscript, yet we feel the cohesive structure in the visualization of Co* is compelling particularly in the mesopelagic demonstrating loss by scavenging, even if its interpretation is not fully understood as of yet. Based on this discussion we feel it is most representative to maintain use of the field values for Co:P in the revision. We point out that though that the choice of any the Co:P value will not affect the trends observed in this visualization, since any change in Co:P will be applied linearly to all Co* values.

The reviewer brings up the matter that dust is not considered much within this manuscript. We have corrected this by incorporating both references and lithogenic corrections of the particulate phase, presented in sections 3.2.2 and 3.3, and presented visually in Figure 11. In almost all cases, and all cases in the upper 400m, the lithogenic component is a minority constituent relative to the biological/authigenic component. Moreover, dust is discussed more extensively in the accompanying Noble et al., BG 2017 manuscript. We have added the following text to demonstrate that pCo is largely biogenic in the study region (although this does not negate the importance of the minority lithogenic phase for Co cycling, a subject beyond the scope of this manuscript):

P9-10: "These oceanic upper water column pCo values here appear to be largely associated with biogenic material: Twining et al. reported pCo in the upper 100m to be largely labile by their leaching methods (Twining et al., 2015), and analysis of the small size fraction from McLane pump samples found the lithogenic component to be a minority component in most samples, particularly in shallower samples (all samples shallower than 400m depth had a minority lithogenic contribution; see Section 3.3 below). Based on these observations and calculations, the assumption of pCo being dominated by biogenic and authigenic components appears reasonable for our stoichiometric discussion, although the observation of the presence of this minority component of lithogenic particulate cobalt in the deep ocean could contribute a slowly dissolving gradual source of dCo in the interior ocean that could be further studied, consistent with prior field and laboratory observations (Noble et al., 2017; Mackey et al., 2015)."

P17: "Lithogenic corrections included here for pCo and previously described for both pCo and pP (Ohnemus and Lam, 2015), show that these elements had minor lithogenic contributions in the North Atlantic particularly in the near surface and typically being a minority contribution at deeper depths, even in the heavily impacted North Atlantic region (Fig. 11a, c, e)."

Responses to specific comments:

Lithium batteries: see response to reviewer #1.

P5. Detection limit values were added, values below 0.02uM were not included from the GA03 dataset.

P7: Dulaquais comparison and reference added, although the value in the Dulaquais paper is 66 not 64.

P8: Regarding the measurement of labile cobalt: we disagree that with the reviewer's statement that it is also complexed and not equivalent to inorganic cobalt. Labile cobalt is indisputably in a much lower detection window that complexed cobalt, and is as a result likely Co(II) in redox state, and could be bound by inorganic or weak organic complexes, as we have documented previously (Saito et al., 2005). Our previous studies have shown one cyanobacterial species is able to take up the strongly bound cobalt, while our and other studies have clearly also shown there to be a inorganic cobalt transporter present in both cyanobacteria and eukaryotic algae as well (Saito et al., 2002, Sunda and Huntsman 1995 and others). We have changed the sentence to reflect this: "The presence of a substantial labile cobalt pool was consistent with the observed high cobalt usage, since labile cobalt is likely highly bioavailable relative to complexed cobalt, particularly for eukaryotic phytoplankton, due to its ability to be taken up through divalent cation transporters (Saito et al., 2002; Saito et al., 2005)."

On the recommendation that we use alternate Redfield ratios: this does not make sense in this context because we are converting laboratory cultures quota data that was grown at replete P conditions, hence standard Redfield is the correct approach.

Moffett and Ho: they only studied surface waters for Mn/Co oxidiation in that study.

Advection is not a process that removes Co from the dissolved phase relative to P (or P to particulate P), hence it is not included here.

dt is used because it reflects changes within a discrete water parcel over time, as is used in 1D models (Johnson et al., 1997; Weber et al., 2007).

Added labels to subfigure 4d, e, and f. Thank you.

Data from Dulaquais et al., 2014 was integrated into Table 1.

Figure 2. The scales are clearly labeled differently. South Atlantic Central Water (SACW) was not one of the water masses characterized in the Ruth et al. 2000, and the similar Saito et al. 2012 South Atlantic OMPA analyses. We have inquired about the possibility of reconstructing the South Atlantic OMPA analyses with similar parameters to the North Atlantic but if it happens it will not be available in time for this revision. The Ross Sea is included as Southern Hemisphere endmember.

Fig 3 caption fixed

Figure 4. Insert made into a panel g

Figure 4. depth was not added as a z parameter since this is illustrated in Fig 3. Figure b and e were integrated further into the discussion.

Figure 5 The reviewer states that "some bottom depths and the depths just above them are circled and it is not in accordance with the 5 point regression being used". The reviewer is pointing out that for the regression to be in the center of 5 vertical points, there should not be regressions at the base of the profiles. For those profiles the bottom depths are actually off scale since this figure uses uniform axes, or the individual two datapoints below each circle are not visible in these small graphs. We have included a table of all regression analyses as supplemental data to allow the reader to examine this.

Figure 6: see discussion above about r values, and added r values in figure 3.

Figure 7: the reviewer asks for dCo data to be included. This data is already in Figure 5.

Figure 10: see discussion above on variability of a mixing vector.

Figure 12: while the r2 values are low, the slopes are negative, which is the intention of the presentation.

Figure 13: z coloring would obscure the current red black coloring scheme.

Figure 14. Isoclines were not added to avoid cluttering the figure.