Response to Reviewer #2 comments on "Radium-228-derived ocean mixing and trace element inputs in the South Atlantic" by Hsieh et al. (bg-2020-377)

Reviewer's comments are shown in black.
***Authors' responses are highlighted in blue.

General Comments The scientific quality of this paper is very good and the quantitative results seem both robust and oceanographically consistent. The calculations and argument are concise and the purpose of the paper is very clear. The comparison of different methods used for radium calculations is useful and provides confidence in the results. However, it seems like the radium side of the paper is significantly more developed than the trace metal side. A little more context and detail could be given to the scientific significance of calculating trace element inputs, and more discussion given to the implications of the calculated trace metal fluxes. Also, the methods of the trace metal calculations, which are an integral part of the paper and several sentences of the abstract, could be moved from the appendix to the main body of the paper. One calculation, the Co/C, Fe/C, and Zn/C export fluxes are noted for being significantly higher than corresponding dissolved inputs. This could be due to the use of laboratory culture metal quota values that were grown under replete conditions. As noted in the paper this region and section has some of the lowest Zn values observed in the euphotic zone from the prior study. Moreover, phytoplankton (and microbes in general) have extraordinary plasticity with respect to metal content, and the lower field and lab scarcity cultures are likely considerably more representative. It would be useful to redo these calculations with metal-carbon values that are more realistic, either using culture studies of phytoplankton grown under scarce rather than replete conditions (e.g. from the relevant Sunda and Huntsman studies) or using particulate metal data from other field expeditions if that data is not available from this section. This would probably help resolve the imbalance in the fluxes compared to the other fluxes calculated and would be more correct in construction of the calculations.

***We thank the reviewer for their positive feedback and constructive comments, particularly for the discussion of trace elements. We respond to all the comments point by point below and explain how we will address the issues in the revised manuscript. Following the reviewer's suggestion, we will also apply a range of reasonable estimates for the TE/C ratios to recalculate the TE removal fluxes in this region.

Specific Comments Introduction: Why are we interested in a calculation of Co, Zn, and Fe flux? Why those elements and not others? Please give more context for trace metals in this region, and the significance of the calculations presented here.

***Iron, zinc and cobalt are known to be essential micronutrients for marine phytoplankton. They all play important roles in the cellular metabolic enzymes and sometimes can collimate primary productivity in different ocean regions. Previous studies have shown that the study region near the 40oS Atlantic is mainly iron limited (Moore et al., 2004; Browning et al., 2014; 2017) and has the lowest reported dissolved zinc concentrations in the global ocean (Wyatt et al., 2014). The requirement for Zn in phytoplankton can be replaced by Co in low Zn areas (Price and Morel, 1990). Thus, knowing the sources and fluxes of these three elements can improve our understanding of the limiting factors for productivity in this study region, one of the most dynamic nutrient regimes in the oceans. We will follow the reviewer's suggestion and add more context in the introduction to highlight the importance of these three elements in this region.

Line 40: Define which metals you'll be talking about (Fe, Zn, Co) here, or at least within the first few paragraphs of the introduction, and why they're important in this region.

***As mentioned above, we will clarify this and add more context in the introduction to highlight the importance of these elements in this region.

Lines 83-91: This paragraph should be moved out of the methods section to the intro- duction or to the results, perhaps as a "hydrographic setting" section in the beginning of the results.

***We agree with the reviewer, but we feel that this paragraph does not fit in either the introduction or the results section. To address this issue, we will thus change the methods section title to "2 Study sites and methods". We will then split the relevant subsection into two, "2.1 Hydrographic setting" and "2.2 Water sampling".

Line 94, 104: Define your trace metal clean technique in more detail. What acids were used and for how long?

***All the trace-metal cleaning procedures follow the GEOTRACES sampling protocols (Cutter et al., 2010). The sample tubing and bottles were rinsed with Milli-Q water and filled with 0.1M HCl for one day. After emptying the acid, the tubing and bottles were rinsed thoroughly with Milli-Q water. The tubing and bottles were also rinsed with open-ocean seawater before sampling. We will add more details in the revised manuscript.

Lines 142-5: Useful analysis of the two methods' uncertainty and error differences. I suggest also performing a pairwise t-test to compare the 226Ra data generated from direct observation and from the Si estimation to determine if the two methods are statistically similar, even with the larger error on the estimated method. This would help convince readers that the methods are comparable.

***Follow the reviewer's suggestion, we have performed a paired t-test and the p-value is 0.55 (> 0.05), which confirms that the measured Ra-226 and the Si-extrapolated Ra-226 data are not statistically significantly different. We will add the t-test results in the revised manuscript.

Line 195: Include sections in the methods to clearly describe how Kz was calculated and the vertical flux of trace metals. They are critical results in your paper and are given prominence in your title and abstract, and their methods should not be buried in your appendix. Perhaps an abbreviated description of the appendix D calculations can be described in the methods, and the full version can be in the appendix.

***We will add more details to describe the calculation of Kz in Section 2.4. As discussed in the response to Reviewer 1 comments, we will summarise the basics of the flux calculations in the main text by adding a new section in the methods (2.5) and keep the calculation details in the appendix.

Lines 211-4: Make this point clearer. You imply but don't clearly state that your results are more similar to Atlantic values than Southern Ocean values. That is an interesting result in this transition zone and should be made more explicit.

***We will rewrite these sentences to clarify this.

Lines 215-22: Expand more on this entire section and convey a more detailed picture of TE distributions in this region. Even if the data has been described elsewhere, this section is too short. Additionally, both Fe and Co do not typically continue to "increase with depth below the mixed layer" because they're scavenged-type elements. Fig. 4 implies that all 3 trace elements increase linearly with depth, which is not necessarily the case beyond the mesopelagic. Give more context to the distribution of all three trace metals here, and qualify that the TEs (specifically Fe and Co) only increase with depth in the upper ocean.

***We will add more context to describe the distribution of these three trace elements in the study region. We will also clarify that the increase in TE concentrations with depth only applies to the upper ocean.

Lines 261-7: Move the description of the Kz calculation to the methods and include the explicit equation for its calculation.

***We will move the Kz calculation to the method section (2.4) and add more details to describe the calculation.

Line 298: As already stated, move at least a brief description of the calculation to the methods section.

***As mentioned above, we will summarise the basics of the flux calculations in the method section (2.5).

Line 317: Briefly expand on low oxygen resulting in higher Co fluxes and provide a citation.

***We will add a few sentences to explain the relationship between oxygen and cobalt fluxes with relevant references.

Lines 322-5: Discuss your findings more than just comparing them to other fluxes. Why do you think they are lower than the other reported Fe fluxes you cite?

***As already mentioned in the manuscript, these high-Fe fluxes are particularly found in regions with river plumes, SGD and the oxygen minimum zone. We will clarify this in the revised manuscript.

Lines 327-30: Same as the above comment for Fe. Why do you think this region has a lower Zn flux? Was this expected?

***There is very limited oceanic Zn flux available for comparison. The only available estimate from the western North Atlantic (Charette et al., 2016) indicates that the cross-shelf Zn fluxes in the South Atlantic are lower than the western North Atlantic. The results may explain why the South Atlantic has the lowest reported Zn concentration in the global oceans. However, it still needs more detailed studies to understand the reasoning behind the different Zn fluxes, which is beyond the scope of this study. We will highlight this in the revised manuscript.

Lines 341-3: This paragraph seems out of place. Either expand on why you're reporting aerosol data and give context for the fluxes (how they compare to the vertical TE fluxes, etc.) or simply move these two sentences to section 4.4 where the numbers are used.

***We will move this paragraph to the beginning of the section, and emphasise that apart from the atmospheric dust deposition, other inputs are still poorly constrained in this region.

Lines 361-72: Specify which of these 3 hypotheses are supported by your results. It sounds like the particles might be a good answer, but it's only mentioned briefly. Expand a bit on what you think is likely going on.

***As discussed in the response to Reviewer 1 comments, the particle hypothesis is based on the observation from Rijkenberg et al. (2014). They have observed high dissolved Fe in the mid depths of this region and suggested that the laterally transported particles from the offshore export waters may release Fe to the upper ocean. However, it is not possible to provide more discussion without further details of the sources and TE concentrations of these particles and the mechanism releasing TEs from these particles. We will highlight this in the revised manuscript.

Figure 3 (lines 745-750) The correlations of metals with salinity is very interesting and worth emphasizing a bit more. Similar observations have been observed on the North American shelf as well observed by Bruland and Franks 1984 and Noble et al. 2017. It could be worthwhile to point out that these observations suggest this is a general feature of Atlantic Western boundaries.

***These observations imply that the distribution of trace elements in the surface ocean is strongly associated with the shelf-water masses in the western boundaries of the Atlantic. We will add a few sentences to highlight this in the revised manuscript.

Bruland, K. W., & Franks, R. P. (1983). Mn, Ni, Cu, Zn and Cd in the western North Atlantic. In Trace metals in sea water (pp. 395-414). Springer, Boston, MA.
Noble, A. E., Ohnemus, D. C., Hawco, N. J., Lam, P. J., & Saito, M. A. (2017). Coastal sources, sinks and strong organic complexation of dissolved cobalt within the US North Atlantic GEOTRACES transect GA03. Biogeosciences, 14(11).

Technical Comments
General: Keep formatting choices consistent. Both 1-D and 1D are used to describe the model, and spaces are inconsistently placed between a number value and its unit. (ex. line 140 uses 100L-1 and line 141 uses 100 L-1.)

***We will check the formatting and ensure consistency in the revised manuscript.

General: Are significant figures to tens of pM for dCo correct?

***This is correct.

Line 102: Define RaDeCC here. It is defined later on line 111, and should be defined at the first use of the abbreviation.

***We will correct this in the revised manuscript.

Line 114: Define MC-ICP-MS here.

***We will define this in the revised manuscript.

Lines 116-7: Briefly clarify why it is necessary to convert from Sr(Ra)SO4 to Sr(Ra)CO3.

***As sulfate is much harder to redissolve back in solutions, the conversion from sulfate to carbonate can improve the efficiency of dissolution. We will clarify this in the revised manuscript.

Line 119: Briefly describe what AG50-X8 and Sr-Spec are. You describe them as ion exchange columns on line 122, but that should be made clearer when they're first mentioned.

***We will clarify these in the revised manuscript.

Lines 126-7: This sentence was already stated on line 114, but without mention of 226Ra. Are you restating the analyses used, or is this a different method? It is not clear.

***This sentence is redundant. We will rewrite and combine it with the next sentence in the revised manuscript.

Line 127: Change Ra-228 to 228Ra to be consistent with the rest of the paper.

***As mentioned above, this sentence will be rewritten and therefore it will not be a problem. However, if an isotope is used in the beginning of a sentence, conventionally the element goes first to avoid beginning the sentence with a number.

Lines 138-40: This is a run on sentence despite the use of parentheses. I suggest removing the parentheses and forming two separate sentences.

***We will rewrite the sentence.

Line 199: "show" not "shows".

***We will correct this.

Line 253: Use a period or semicolon between "waters" and "therefore" to avoid the run on sentence.

***We will make the change.

Line 283: Figure 7 only shows depth, not sigma-t as this line states. Rephrase this sentence or relabel the figure to show the density ranges instead of depths.

***We will rephrase the sentence.

Line 297: Is the shelf 228fluxes point a fourth part of this list, or is it a separate point from the 3 approaches list? If it's part of the list, label it as (4). If it's a separate point, put it in its own sentence.

***The shelf 228Ra flux is only used for the TE/228Ra-ratio-derived TE fluxes. The sentence will be rewritten to clarify this in the revised manuscript.

Lines 309-10: Confusing sentence – nothing is being compared here. Rephrase.

***The sentence will be rephrased to avoid confusion.

Figures: The figures here do a good job of supporting the data and conclusion of the work, but they should be edited for readability. In general, do not rely on color coded axes to label subplots (especially for colorblind people), and include axis labels with units clearly next to the axis number scale. Avoid breaking up the axis label and the axis unit. Also, consider putting some of the regression equations in a table or in the appendix, particularly if they're not referenced in the text.

***We will check and correct these problems in the revised manuscript.

Fig 1: The stations numbers are crammed into the top plot b and are mostly illegible. The Salinity z-axis label is also in a strange location. I suggest leaving more space between plots a and b to include this information above figure b.

***We will make the changes.

Fig 4: Add y-axis labels for depth. Also, the x-axis labels are difficult to find. Is there a way to make the metal label clearer for each row?

***We will add the depth label and clarify the metal label for each row.

Fig 6: The x-axis labels for the top row of plots are not very readable here. The 228Ra label should be next to its units and ideally should be above the axis like the labels for the bottom row of plots. The subtle color coding for sigma-t and T is also confusing.

***We will remake the figure to avoid these problems.

***Other references cited in the response:

Browning et al. (2014) Nutrient regimes control phytoplankton ecophysiology in the South Atlantic, Biogeosciences, 11, 463-479.

Browning et al. (2017) Iron limitation of microbial phosphorus acquisition in the tropical North Atlantic, Nature Communications, 8, 15465.

Charette et al. (2016) Coastal ocean and shelf-sea biogeochemical cycling of trace elements and isotopes: lessons learned from GEOTRACES, Phil. Trans. R. Soc. A, 374, 20160076.

Cutter et al. (2010) Sampling and sample-handling protocols for GEOTRACES cruises. http://www.geotraces.org/libraries/documents/Intercalibration/Cookbook.pdf

Moore et al. (2004) Upper ocean ecosystem dynamics and iron cycling in a global three-dimensional model. Global Biogeochemical Cycles, 18, GB4028.

Price and More (1990) Cadmium and cobalt substitution for zinc in a marine diatom, Nature, 344, 658-660.

Rijkenberg et al. (2014) The distribution of dissolved iron in the west Atlantic Ocean, PLoS One, 9, e101323.

Wyatt et al. (2014) Biogeochemical cycling of dissolved zinc along the GEOTRACES South Atlantic transect GA10 at 40°S, Global Biogeochem. Cy., 28, 44-56.