Response to Referee 1

We thank Referee 1 for the helpful comments. We will address all changes in the revised manuscript as detailed in our responses below. The referee comments are in black and their line numbers refer to the original submitted manuscript. Our responses are in blue text.

We want to note that some of the reviewer's comments may pertain to an original draft of the manuscript which has already been revised. We have tried to address all comments, but in some cases, we do not see what the reviewer is talking about. In our response, we will only be referring to the version that is currently available on the BG website.

We also want to note that we now have submitted a companion paper to this special issue that is specifically about the particulate organic carbon (POC) export using the $^{210}Po/^{210}Pb$ technique.

- Specific Comments: One unfortunate aspect of the paper is that it fails to model the data in the context of biogenic carbon flux, the primary strength of the nuclide pair. Perhaps the organic carbon data are missing, or awaiting a more complete synthesis with other nuclides such as 234-Th, as done admirably before by the UAB lab group.

We understand the reviewer's concern. In fact, we have submitted two manuscripts to this special issue. In the manuscript reviewed here we discuss the general distribution of $^{210}Po$ and $^{210}Pb$ activity along the GEOVIDE transect. The second manuscript entitled "The export flux of particulate organic carbon derived from $^{210}Po/^{210}Pb$ disequilibria along the North Atlantic GEOTRACES GA01 (GEOVIDE) transect" addresses the POC export fluxes. In the second paper, we have calculated the POC fluxes using the export flux of $^{210}Po$ and the POC/$^{210}Po$ ratio in total (> 1 µm) particles and compared the estimates to those obtained using the $^{234}Th/^{238}U$ proxy.

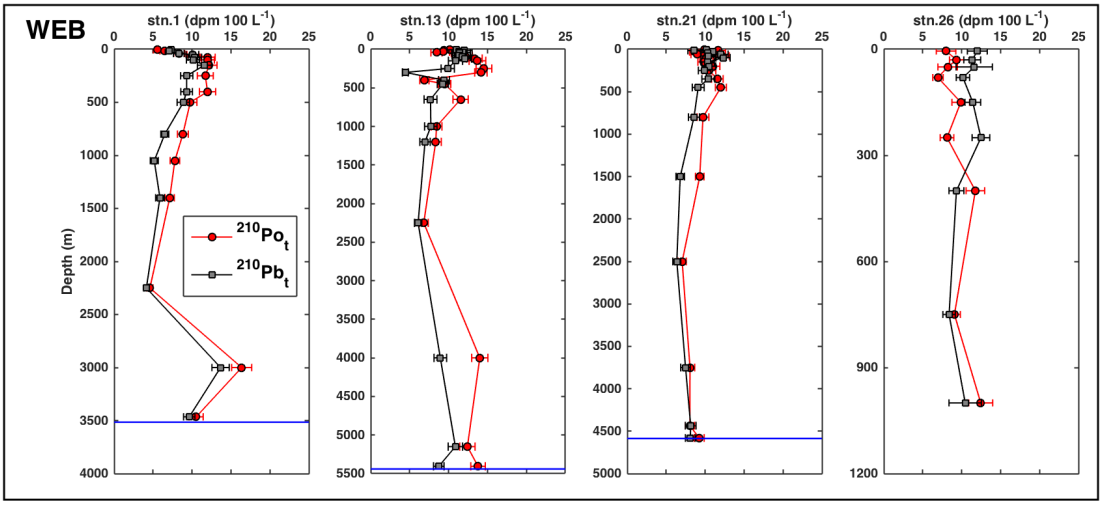
Technical Issues:

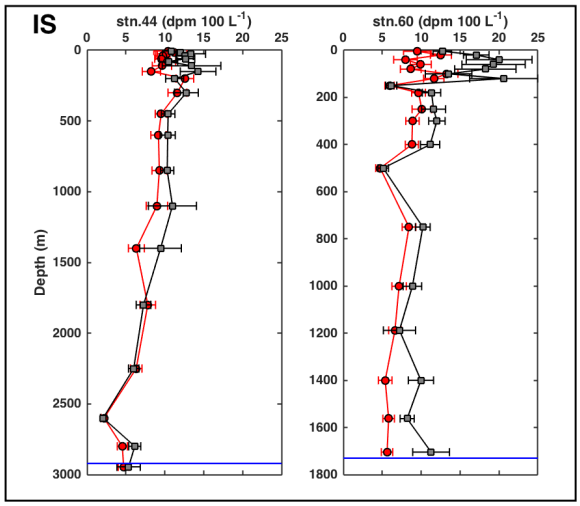
1. Introduction It is noted that there is significant benthic disequilibrium (210-Po deficiency) well below the euphotic zone, indeed significantly below the main thermocline at times (e.g. 4000 meters at Station 13; 1400 meters at station 60). This dilemma and benthic consequences has been discussed in the recent literature (Rigaud, et al., 2014). Page 3: As such, maybe the literature citations in the introduction that need to be updated for the current millennium!

We agree that $^{210}$Po deficits at depth can be problematic in interpretation, but others have associated them with nepheloid layers or other forms of suspended sediment near the bottom (e.g. Hu et al., 2014; Wei et al., 2014; Rigaud et al., 2015). Mid-depth deficits are more mysterious and have still not really been adequately resolved (Kim, 2001; Church et al., 2012; Rigaud et al., 2015).

We acknowledge that there were deficits in the deep waters at stations 60, and 64. However, we have almost no particle data for those depths so cannot address that in this paper.

In Fig. 2, total $^{210}$Po and total $^{210}$Pb activity were plotted as red circles and black squares, respectively. There were indeed $^{210}$Po deficits below 1400 m at station 60 but at 4000 m at station 13 there was a $^{210}$Po excess rather than a deficit. Please see the following profiles for stations 13 and 60.






We agree that the literature citations on Page 3 L55-57 need to be updated. We will add in some recent references (highlighted in bold font) as the following:

"The distribution of $^{210}$Po and $^{210}$Pb has been widely measured over the last several decades in the Atlantic (e.g. Bacon et al., 1976; Sarin et al., 1999; **Rigaud et al., 2015**), Pacific (e.g. Nozaki and Tsunogai, 1976; Murray et al., 2005; Verdeny et al., 2008), Indian (e.g. Cochran et al., 1983; Sarin et al., 1994; **Subha Anand et al., 2017**), Arctic (e.g. Moore and Smith, 1986; **He et al., 2015; Roca-Martí et al., 2016**) and Southern Oceans (e.g. Shimmield et al., 1995; Friedrich and Rutgers van der Loeff, 2002)"

2. Methods Page 4: What is meant by "Xlarge" station (26), as the number of depths are less than others?

Station naming depended on the number of casts that were conducted: XLarge stations had five casts while Super stations had > 5 casts. Because of the fewer casts, we did indeed sample at fewer depths at Station 26 than at the other stations.

3. Page 5: Is six hours sufficient for equilibration, or were there previous tests performed to verify this?' What was the time lag between sample processing on board, and nuclide separation in the lab on shore, unless both were done on board? This can be important as reviewed in Rigaud, et al. 2013. Evidently this is reflected in the data reported in

supplemental tables, although there are not errors assigned to the nuclide ratios in Table 2.

Thank you for this point. Six hours for isotope equilibration was a mistake. In fact we waited for more than 12 hours for isotopic equilibrium between $^{210}$Po and $^{209}$Po, which is recommended in RiO5 Cookbook (https://cmer.whoi.edu/wp-content/uploads/2018/01/15-Po-Pb-210-in-sewater_Co-APDC.pdf) and suggested in Rigaud et al. (2013). We will revise the manuscript accordingly.

We did our best to minimize delays in sample processing. The time elapsed between sampling and nuclide separation (first plating) was 50 and 68 days on average for the seawater samples processed at UAB and QC, respectively, and 58 and 44 days on average for the particulate samples processed at UAB and QC, respectively. This was unavoidable as the cruise was long. However, all this is taken into account into the corrections and calculations, which were performed as described in Rigaud et al. (2013).

The errors for the total particulate $^{210}$Po/$^{210}$Pb activity ratio ($^{210}Po_p/^{210}Pb_p$) are now added in Table 2 as follows:

Table 2. The compilation of total particulate $^{210}Po/^{210}Pb$ activity ratios ($^{210}Po_p/^{210}Pb_p$) averaged in the upper 200 m, including this study.

| Region | | Sampling Method | Date | Size (μm) | Depth (m) | $^{210}Po_p/^{210}Pb_p$ | Reference |
|---|---|---|---|---|---|---|---|
| Arctic | CESAR | *In-situ* pump | Apr – May 83 | > 0.45 | 2-200 | 1.2 ± 0.7 | (Moore and Smith, 1986) |
| | Arctic (ARK-XXII/2) | Niskin bottle | Jul-Sep 07 | > 1 | 10-200 | 0.50 ± 0.20 | (Friedrich, 2011) |
| | Chukchi Shelf | Niskin bottle | Jul-Sep 10 | > 0.45 | 0-90 | 0.37 ± 0.10 | (He et al., 2015) |
| Atlantic | F.S. Meteor | Niskin bottle | Nov-Dec 73 | > 0.4 | 0-200 | 3.1 ± 1.4 | (Bacon, 1977) |
| | Cariaco Trench | Niskin bottle | Dec 73 | > 0.4 | 0-200 | 1.4 ± 0.6 | (Bacon et al., 1980a) |
| | Labrador (R/V Knorr) | Niskin bottle | Jun 75 | > 0.4 | 0-100 | 3.9 ± 1.5 | (Bacon et al., 1980b) |
| | South of New England | Niskin bottle | Jul 80 | > 0.45 | 4-200 | 1.8 ± 0.8 | (Bacon et al., 1988) |
| | N. Atlantic (BOFS) | Niskin bottle | May-Jun 89, 90 | > 0.45 | 0-150 | 6.0 ± 4.5 | (BODC et al., 2016) |
| | South-equa. Atlantic | Niskin bottle | May-Jun 96 | > 0.7 | 10-200 | 1.3 ± 1.1 | (Sarin et al., 1999) |
| | BATS | Go-Flo bottle | Oct 96 | > 0.45 | 0-200 | 3.7 ± 3.2 | (Kim and Church, 2001) |
| | N. Atlantic (GA03) | *In-situ* pump | Oct-Nov 10, Nov-Dec 11 | > 0.8 | 30-200 | 1.5 ± 0.5 | (Rigaud et al., 2015) |
| | N. Atlantic (GA01) | *In-situ* pump | May-Jun 14 | > 1 | 8-200 | 1.4 ± 0.3 | This study |
| Pacific | North Pacific | Niskin bottle | Nov 73 | > 0.4 | 10-150 | 8.5 ± 5.7 | (Bacon et al., 1976) |
| | W. Pacific (FR05/92) | Niskin bottle | Jul 92 | > 0.45 | 0-200 | 1.3 ± 1.0 | (Towler, 2003) |
| | Equa. Pacific | Go-Flo bottle | Aug-Sept 92 | > 0.45 or 0.5 | 0-200 | 5.1 ± 1.2 | (Murray et al., 2005) |
| | W. Pacific (FR08/93) | Niskin bottle | Nov 93 | > 0.45 | 0-200 | 16 ± 4 | (Towler, 2013) |
| | W. Pacific (FR07/97) | Niskin bottle | Aug 97 | > 0.45 | 0-200 | 7.2 ± 1.5 | (Peck and Smith, 2002) |
| | Aleutian Basin | Niskin bottle | Jul-Aug 08 | > 0.2 | 0-200 | 1.9 ± 3.0 | (Hu et al., 2014) |
| | E. Pacific (GP16) | *In-situ* pump | Oct-Dec 13 | > 1 | 15-200 | 2.4 ± 0.6 | unpublished |
| Antarctic | S. Ocean (ANT-X/6) | Niskin bottle | Oct-Nov 92 | > 0.45 | 20-200 | 3.0 ± 1.4 | (Smetacek et al., 1997) |
| | Bellingshausen Sea | Go-Flo bottle | Nov-Dec 92 | > 0.45 | 0-100 | 14 ± 11 | (Shimmield et al., 1995) |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | S. Ocean (ANT-XXIV/3) | Niskin bottle | Feb - Apr 08 | > 0.45 | 25-200 | 1.3 ± 0.9 | (Friedrich et al., 2011) |
| Margin Sea | S. China Sea | Go-Flo bottle | Jan-Oct 07, May 08 | > 0.45 | 0-200 | 1.7 ± 1.1 | (Wei et al., 2014) |
| | W. Taiwan | Go-Flo bottle | Apr 07 | > 0.45 | 8-25 | 0.85 ± 0.12 | (Wei et al., 2012) |
| | Yellow Sea | Niskin bottle | Feb 93 | > 0.7 | 0-100 | 0.88 ± 0.08 | (Hong et al., 1999) |
| | Mediterranean Sea | Sediment trap | Mar-Jun 03 | | 200 | 4.5 ± 1.0 | (Stewart et al., 2007) |

4. Page 6: Who are the “Planquette group”?

Helene Planquette Group, University of Brest, co-authors in this issue.

5. Results Page 7: As noted above, stations 13 and 60 appear to have total 210-Po deficiency at depth (Fig. 2), not excess.

Please see our response to Technical Issue 1.

6. Page 8: Increase in activity with depth for both nuclides is not evident in Figs. 2 and 3, rather decrease.

Figures 2 and 3 are the profiles of total radionuclide activities ($^{210}Po_t$, $^{210}Pb_t$) from surface to bottom and from surface to 250 m, respectively.

In the original manuscript Page 8 lines 214-216: “The vertical profiles of $^{210}Pb_s$ were generally similar to those of $^{210}Po_s$, with relatively high activity in the surface, lower activity in the subsurface and increasing activity with depth;” $^{210}Pb_s$ and $^{210}Po_s$ refer to particulate activity in the small size fraction (not totals as in Figs 2 and 3). This data was shown only in supplementary Table S2, but we realize that it should be included in the paper and have added figures.

We will include the vertical profiles of small and large particulate radionuclide activity as the following:

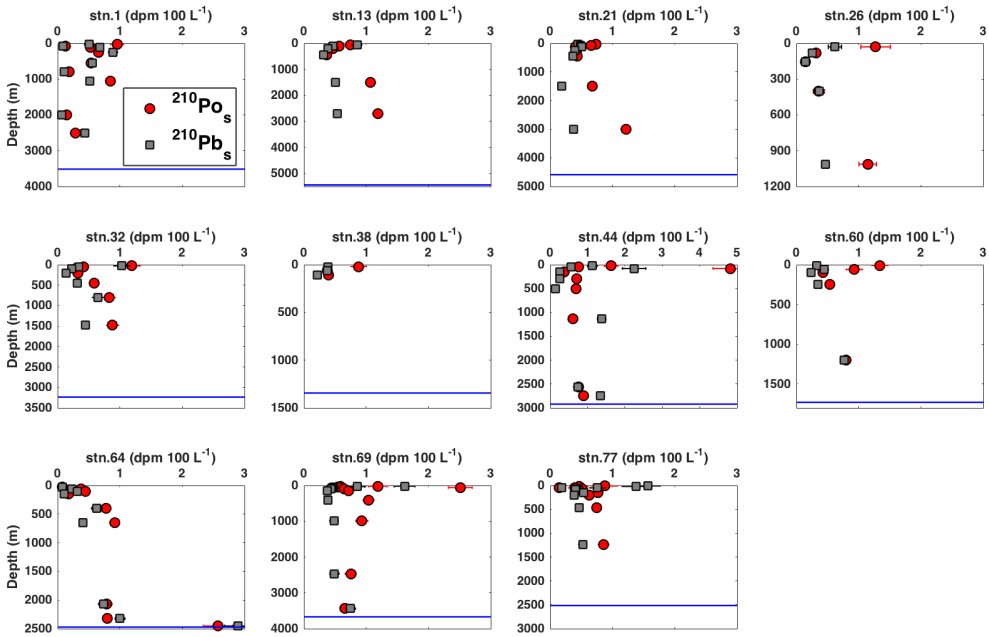



Fig. 4. Vertical profiles of the particulate $^{210}$Po and $^{210}$Pb activity in the small size fraction (1-53 µm, $^{210}Po_s$, $^{210}Pb_s$). Note the different depth scales for the various stations and that the activity scale at Station 44 differs from the scale of all other stations. The horizontal blue line represents the bottom depth at that station.

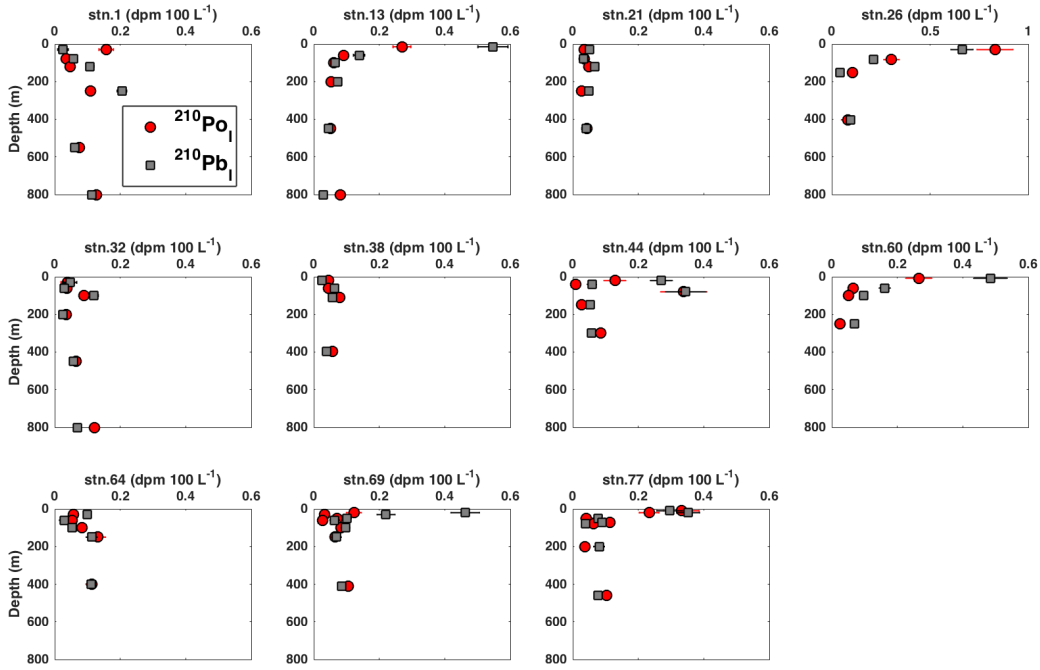


Fig. 5. The vertical profiles of the particulate $^{210}$Po and $^{210}$Pb activity in the large size fraction (> 53 µm, $^{210}Po_l$, $^{210}Pb_l$) in the top 800 m. Note that the activity scale at Station 26 differs from the scale at all other stations.

Discussion Page 10: Usually in the far North Atlantic, 210-Pb association with aerosol dust is not as evident in the east, rather alternative fresh water sources (e.g. precipitation) as noted in the west.

We agree. In the submitted manuscript, we acknowledged the possible inputs of $^{210}$Pb from freshwater (e.g. sea ice processes and meteoric water) in the high latitude North Atlantic, in particular near the Greenland shelf. Nonetheless, we removed the word "unexpected" from the last sentence of this paragraph on Line 281.

7. Page 11. The lithogenic source of a depleted 210-Po/210-Pb ratio should only be evident if the atmospheric scavenging was in the form of precipitation. Alternatively or as with lithogenic particles from the continental margin, the 210-Po has been preferentially extracted lately in fecal pellets by organisms.

We find this statement a bit confusing and are not sure how to best address it.

Near the coast, most of the lithogenic particles are terrestrial/riverine particles with a small contribution from aerosols. Aerosols have a very low $^{210}$Po/$^{210}$Pb AR (< 0.2, Baskaran, 2011) due to the short residence time of $^{210}$Pb in the atmosphere (e.g. Moore et al., 1974; Turekian et al., 1977). For the lithogenic particles sourced from land/river, the particulate $^{210}$Po depletion is more related to the nature of those particles that may preferentially adsorb $^{210}$Pb vs. $^{210}$Po as opposed to the patterns in organic materials (e.g. Fisher et al., 1983; Stewart et al., 2005).

8. Page 12: The alternative scenario is noted here at the end of section 4.2. As such, might there be a corresponding dissolved ration greater than one?

Yes, we have looked at this relationship and will address it in much greater detail in an upcoming manuscript entirely about this topic (data presented at the Ocean Sciences Meeting, February 2018). In this study, there were a total of 13 depths where the particulate $^{210}Po/^{210}Pb$ activity ratio was lower than 1 and 8 of these depths also had a dissolved $^{210}Po/^{210}Pb$ activity ratio lower than 1.

9. Page 13: The negative relationship between AOU and 210-Po/210-Pb is not very strong.

We agree that both negative and positive linear relationships between total particulate $^{210}Po/^{210}Pb$ AR and AOU are not very strong, with $R^2$ as 0.5 and 0.4, respectively. Nonetheless, the p-value for both linear relationships is below the significance threshold of 0.05. It appears that the negative relationship was stronger than the positive relationship in terms of $R^2$ and p-value. If we had more data available (both particulate $^{210}Po$ and $^{210}Pb$ activity and AOU), it would perhaps be possible to observe stronger relationships. This is a topic that will be explored in the future and we thought it was helpful to show the relationships while providing possible mechanisms to explain them.

10. Page 14: Line 387 appears not to be clearly expressed indeed!

We agree and will add the following sentence to make it clearer:

"As claimed previously in Tang et al. (2017), $K_d$(Po) is complicated because it appears to reflect both the surface adsorption and potential bioaccumulation."

11. Conclusion Page 15: The impact of a terrestrial origin on the 210-Po/210Pb ratio less that unity might indeed be born out in the Arctic basin during summer seasons of strong biogenic processing. Maybe there is evidence in the recent GEOTRACES cruises on time scales of several months conclusive with that of the grand-daughter/parent nuclide pair?

There are two recent GEOTRACES Arctic cruises (GN01 and GN04) in 2015 which both have sampled for $^{210}Po$ and $^{210}Pb$ activity measurements. Unfortunately, the data is not yet available.

12. Figure Captions 4) …bloom defines the date when the next bloom began.

Corrected.

13. Figure Caption 5) The black and blue colored circles are not well distinguished.

The color code in Figure 5 is now modified. Please see below.

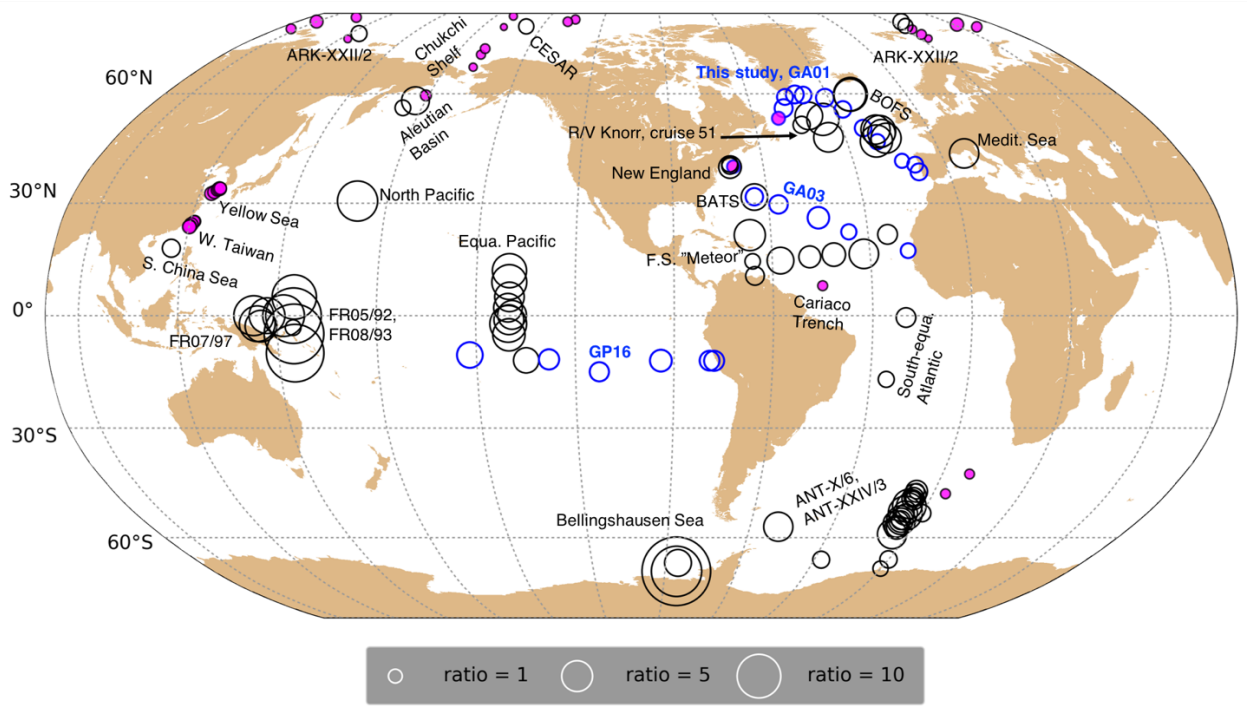



Fig. 8. Comparison of particulate $^{210}Po/^{210}Pb$ activity ratios in the upper 200 m from this study and 20 previous studies (references in Table 2). Information about the study site, sampling date, method, and particle size of each study are shown in Table 2. The black circles represent data from previous studies while the blue circles are the results from samples analyzed at QC from three recent GEOTRACES transects (GA03, GP16, and this study, GA01 GEOVIDE). The filled magenta and open circles indicate activity ratios lower and higher than 1, respectively.

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