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.

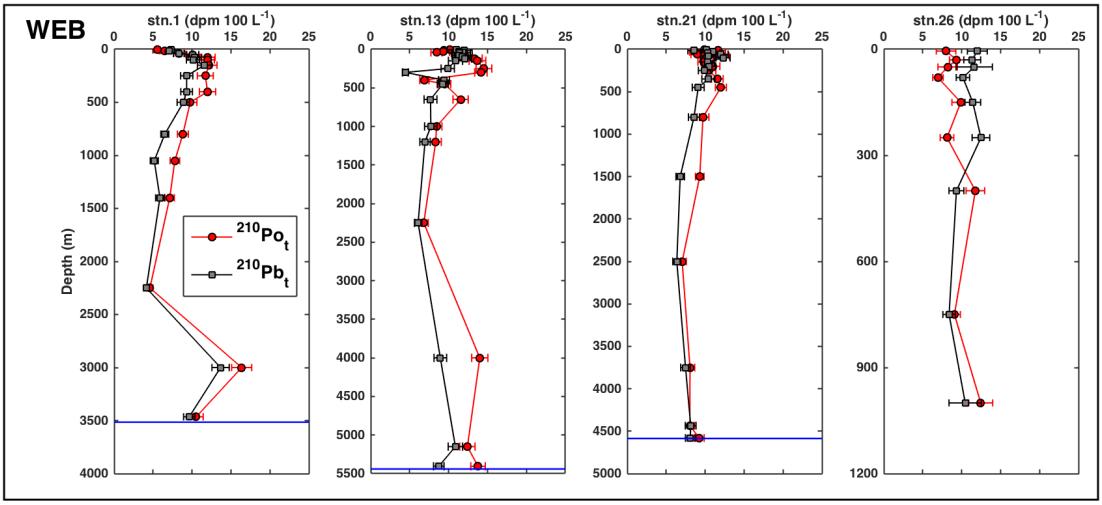
Technical Issues:

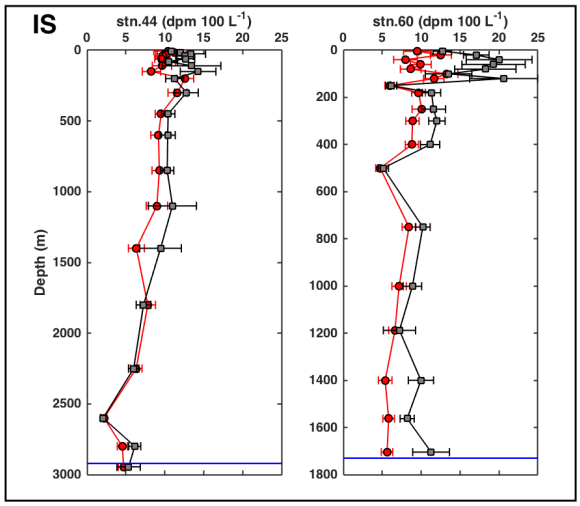
1. Introduction It is noted that there is significant benthic disequilibrium (210-Po deficiency) well below the euphotic zone, indeed significantly below the main thermocline at times (e.g. 4000 meters at Station 13; 1400 meters at station 60). This dilemma and benthic consequences has been discussed 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:

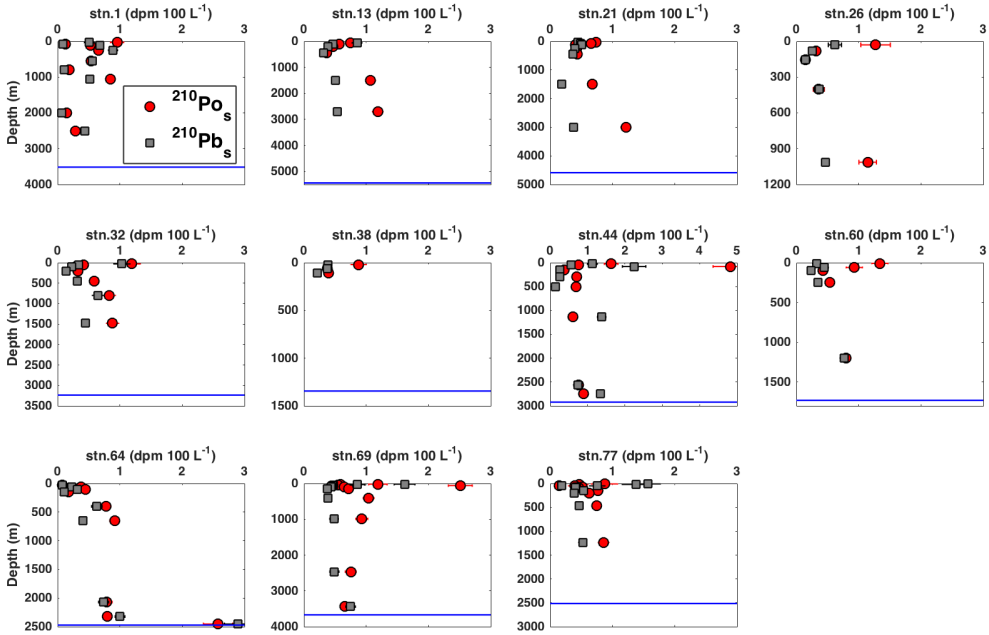


Fig. 4. Vertical profiles of the particulate ^{210}Po and ^{210}Pb activity in the small size fraction (1-53 µm, $^{210}Po_s$, $^{210}Pb_s$). Note the different depth scales for the various stations and that the activity scale at Station 44 differs from the scale of all other stations. The horizontal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

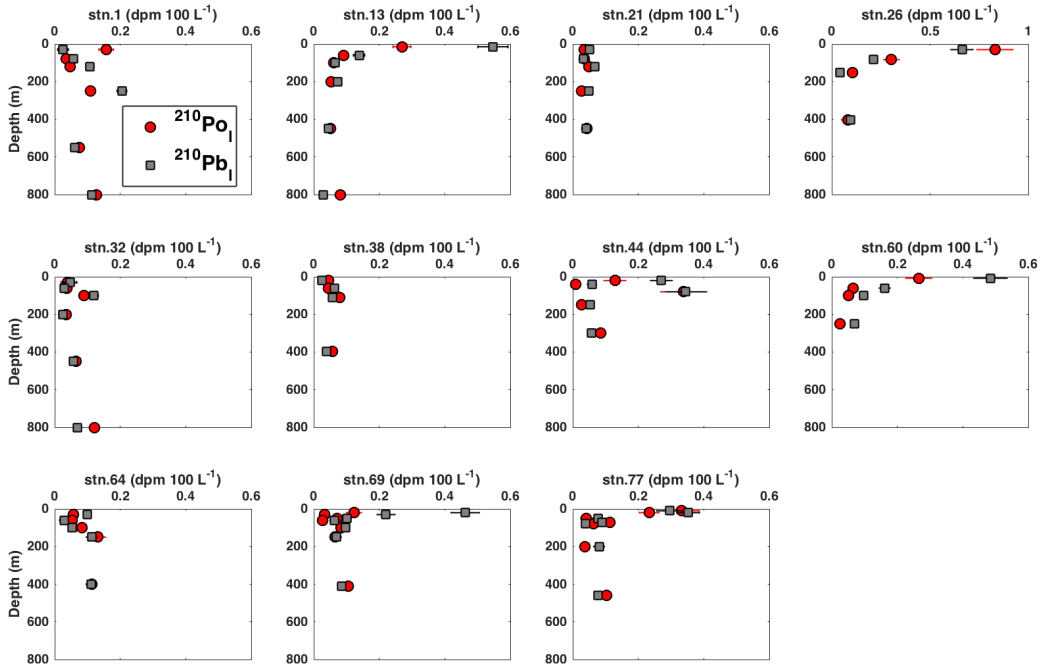


Fig. 5. The vertical profiles of the particulate ^{210}Po and ^{210}Pb activity in the large size fraction (> 53 µm, $^{210}Po_l$, $^{210}Pb_l$) in the top 800 m. Note that the activity scale at Station 26 differs from the scale at all other stations.

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.

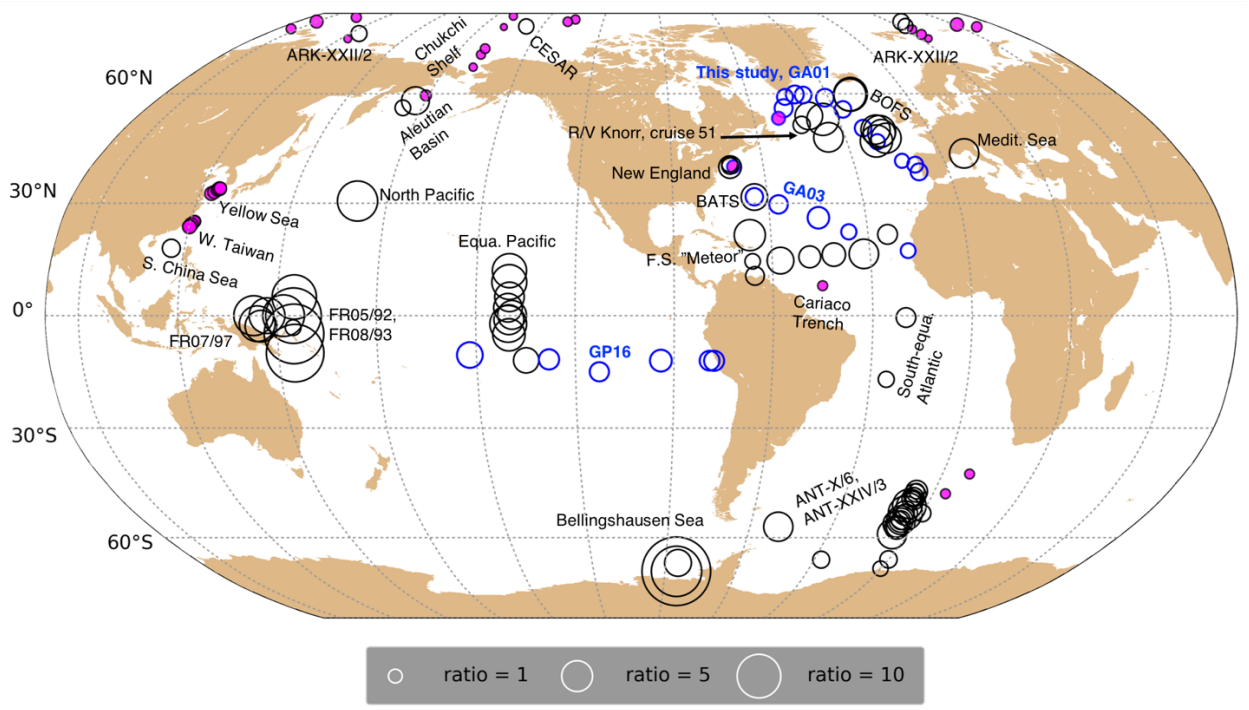


Fig. 8. Comparison of particulate $^{210}Po/^{210}Pb$ activity ratios in the upper 200 m from this study and 20 previous studies (references in Table 2). Information about the study site, sampling date, method, and particle size of each study are shown in Table 2. The black circles represent data from 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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Response to Referee 2

We thank Referee 2 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.

Reviewer Recommendation and Comments for Manuscript BG-2018-210

General comments

The manuscript reports on total, small particles and large particles activity of 210Po and 210Pb nuclides along the North Atlantic GEOTRACES GA01 (GEOVIDE) cruise. The paper is well written, well structured and I believe that such measurements in this areas are essential for the scientific understanding of TEI's and biogenic elements in the global ocean. The approach is very good and the compilation of many other joined data (AOU, PP, SPM, chlorophyll, …) is essential to reach this goal. In addition there is a huge effort to include this new dataset with previous ones in order to get a better view of the processes controlling the behaviors of 210Po and 210Pb at a larger scale. Finally I found very interesting news findings that emerge from a new way to confront this 210Po and 210Pb dataset to other variables (comparison with chlorophyll-a from satellite-based data, AOU, …) that merit to be published.

However I found some questioning points that need to be addressed:

1. the splitting of the samples between two different labs with two methods that differ in some points is very surprising. Some practical reasons can certainly explain this

procedure but they are not mentioned. The reader need to be sure that the results can be compared. Especially since there are distinct features that can be seen between the samples from the two labs and that a part of the discussion relies on such differences.

Thank you for your suggestion. We agree that there are some differences in the procedure between the two labs. We will add explanation in the text. Please see our responses to Specific comments 13 and 14.

2. the last section of the discussion about the sorption, distribution coefficient and implication for particles and POC export very speculative. This is embarrassing as this appears in the abstract and the conclusion as the most important finding of the study while there are other findings much more robust that are not presented in that way.

There may be a misunderstanding, but we don't see POC export in the current abstract. We have submitted a companion paper to this special issue that is specifically about POC export, unlike this paper. We will remove the reference to POC export and rephrase the last section of the discussion to support our observations of ^{210}Po and ^{210}Pb distributions. Please see our response to Specific comment 47.

3. the presentation of the context in the introduction and the state of the art about 210Po and 210Pb isotopes in the ocean is a little bit weak and I think the importance of such measurement in this area should be specifically strengthened.

Thank you for your suggestion. We will edit the Introduction section by adding more rationale for the GEOVIDE section and strengthening the objective section. Please see our responses to the Specific comments 4 and 5.

Consequently, I believe this paper must be published when these points will be addressed.

Thank you for your positive evaluation. Please see our responses to the specific comments below.

Specific comments:

Title:

1. The part of the title "partitioning between the dissolved and particles phase" is maybe not really appropriate as the most important discussions in the paper is about the processes explaining the variations in the 210Po/210Pb activity ratio within each phase (i.e., total and small/large particles).

We agree and will change the title to "Distributions of total and size-fractionated particulate ^{210}Po and ^{210}Pb activities along the North Atlantic GEOTRACES GA01 (GEOVIDE) transect".

Abstract

2. P2, L22-23: this was not shown in the manuscript

We agree that we didn't mention it in the original manuscript.

We will add the sentence of "The average values of K_d(Po) was 1.6 times of those of K_d(Pb) in both small and total particulate phases, suggesting a higher affinity with particles for ^{210}Po with respect to ^{210}Pb, which is commonly observed in the global ocean (Bacon et al., 1988; Hong et al., 1999; Masqué et al., 2002; Wei et al., 2014; Tang et al., 2017)." on L399. We will keep this in the Abstract after the addition.

Introduction

3. P3, L42: "seventh repetition of the OVIDE section": please precise what is the OVIDE section/program?

OVIDE is an acronym of "Observatoire de la variabilité interannuelle et décennale en Atlantique Nord," and this section covers Portugal to Greenland. This will be clearly explained in the summary paper of this special issue (Sarthou et al., in review).

4. P3, L44-46: please give a short summary on the hydrological properties on the area.
   P3, L47-49: you should illustrate what is this expected "mixture of complex …" and why this section may present a special opportunity.

We agree that we did not provide detailed information on the hydrological properties of the study area nor the rationale for the cruise track. There will be multiple papers in this special issue and in other journals specifically describing the hydrographic and physical characteristics, justification of the GEOVIDE cruise track, and the sampling strategies (e.g. García-Ibáñez et al., 2015; Benetti et al., 2017; García-Ibáñez et al., 2018; Zunino et al., 2018; Sarthou et al., in review). We therefore will only add some information about the section as the following:

"The major goal of the international GEOTRACES program is to characterize the distributions of trace elements and isotopes (TEIs) in the ocean on a global scale, and to identify and quantify processes that control these distributions (GEOTRACES Planning Group, 2006). The GEOVIDE section was a contribution of the French GEOTRACES program to this global program in the subpolar North Atlantic. The GEOVIDE GA01 cruise was carried out in 2014 in the North Atlantic and consisted of two sections: a section along the OVIDE (Observatoire de la variabilité interannuelle et décennale en Atlantique Nord) line between Lisbon (Portugal) and Cape Farewell (southern tip of Greenland), and a Cape Farewell to St. John's (Canada) section across the Labrador Sea (Fig. 1). Since 2002, the OVIDE section has been occupied biennially to collect physical and biogeochemical data (Mercier et al., 2015). The knowledge of the currents, water masses, and biogeochemical provinces gained from the previous OVIDE campaigns enabled the optimal strategy for TEIs sampling and provided help for the interpretation of the distribution of TEIs in the subpolar North Atlantic (García-Ibáñez et al., 2015). In addition to the OVIDE line, the Labrador Sea section provided a unique opportunity to study TEIs distributions along the boundary current of the western North Atlantic subpolar gyre (Sarthou et al., in review)."

5. P4, L72-78: I found this objectives section disappointing and clearly not ambitious enough with respect to the dataset compiled and presented in this paper. I suggest the authors to strengthen this part.

We agree with the reviewer's comments and will rewrite the objectives section as:

"In this work, we describe the distributions of total and size-fractionated particulate ^{210}Po and ^{210}Pb activity along the GEOVIDE cruise in the North Atlantic. These data are a significant

contribution to the high-latitude North Atlantic ^{210}Po and ^{210}Pb activity data set. We present a compilation of particulate ^{210}Po/^{210}Pb activity ratios (AR) from previous studies in the global ocean and the results are discussed in regards to the aging of water and biochemical processes. We also describe the relationship among small particles, adsorption, and scavenging of radionuclides. These results lead to recommendations for the estimation of particulate organic carbon export flux based on the ^{210}Po/^{210}Pb disequilibrium, a topic that is covered in a companion paper (Tang et al., submitted)."

Methods

6. P5, L104: Please correct the sentence to avoid confusion: what was transferred into a clean bottle? The filters? The filtrate?

The filter was placed into a clean falcon tube. We will rephrase the sentence as: "Samples were filtered through a 0.45 µm membrane filter and the filters with the precipitate were placed into falcon tubes, sealed with parafilm, and stored in double-bags."

7. P5, L107: is the "Stewart laboratory" the official name of the laboratory?

The lab doesn't have an official name. Because G. Stewart is the investigator of this lab at Queens College (QC), we used "Stewart Laboratory" in the text. We now will use QC in the revised manuscript.

8. P5, L107-108: why this splitting procedure of the sample? The reader need to know why this splitting procedure allow to "ensure higher counting statistic in the samples". Did the laboratory performed intercalibration experiments?

We will answer the question in the text as follows: "As the delay between sample collection and first Po plating increases, the uncertainty of the calculated ^{210}Po activity also increases. In addition, it is necessary to balance counting periods with the number of samples as the uncertainty due to alpha spectrometry counting decreases by increasing the counting time. To limit the delay between sampling and processing and to ensure higher counting statistics by having more alpha spectrometers devoted to this project, sample processing and analyses were

split between Universitat Autònoma de Barcelona (UAB) (samples from stations 1, 13, and 21) and Queens College (QC) (stations 26, 32, 38, 44, 60, 69, and 77)."

Unfortunately, there wasn't enough material to perform an intercalibration experiment.

9. P5, L110: Please correct the sentence to avoid confusion: the filter was not evaporated to dryness.

We will change the sentence to "Briefly, the filters were digested into a solution of concentrated HNO_3 and HCl, and after the solution was evaporated to dryness, the samples were recovered in 1M and 0.5 M HCl solution at UAB and QC, respectively (a 0.5-2 M HCl solution is recommended, Rigaud et al., 2013)."

10. P5, L110: Remove "eventually"

Done. Please see the previous response.

11. P5, L112:what weak acid solution?

We now write this as "1 M/ 0.5 M HCl solution".

12. P5, L120: write "to determine Pb recovery" instead of "to determine sample recovery".

Done.

13. P5, L125-127: why this difference between the two labs?

The higher uncertainties for the samples processed at QC were due to additional corrections on the 1st stable Pb recovery. We will explain this in the text as:

"The activities of ^{210}Po and ^{210}Pb at the time of collection were determined by a series of corrections, including nuclide decay, ingrowth, chemical recoveries, detector backgrounds, and blank contamination following the methods in Rigaud et al. (2013). The activity uncertainties from UAB were on average 8% for both ^{210}Po and ^{210}Pb activity, while the QC uncertainties were on average 13% for ^{210}Po activity and 16% for ^{210}Pb activity. The greater uncertainties of

^{210}Po and ^{210}Pb activities in the samples processed at QC were due to the longer delay between sampling and first plating (68 vs. 50 d) and higher uncertainties in the determination of the recovery of lead."

14. P6, L137: These two different digestion procedures may give different results? Please explain if tests were carried out. Are the data from the two groups comparables?

We agree that there were different digestion procedures (with or without HF), and we didn't run comparisons and need to rely on both labs working well.

15. P6, L144: what is the Planquette group?

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

16. P6, L144: is this sentence correct: "the material on the balance of the screens and filters"?

The sentence will be rephrased as "The Helene Planquette group (University of Brest) collected subsamples from the same screens and filters that were sampled previously for radionuclides to determine major phase composition (particulate organic matter (POM), lithogenic material, calcium carbonate ($CaCO_3$), opal, $Fe(OH)_3$, and MnO_2) (references therein Lam et al., 2015)."

17. P6, L148-149: if the method is the same as described by Lam et al. 2015, I suggest to remove the Lemaitre et al. in prep. reference if it is not published at the time of the publication of this paper. Same comment for other reference in prep. in the manuscript.

Thank you for your suggestion. We will remove the references in prep. from the manuscript but keep the references submitted or in review as the journal suggests.

18. P6, L158: what is the Dehairs group?

Frank Dehairs group, Vrije Universiteit Brussel, co-authors in this special issue.

19. P6, L157-164: a little bit more details is needed here: how the photometric conditions was applied on deck? I guess that 13C was spike before the incubation? …

Yes, the sample was spiked with $NaH^{13}CO_3$ before the incubation. More details on the experimental procedure were added in the text as: "The seawater was then spiked with 3 mL of a $NaH^{13}CO_3$ solution (200 mmol L^{-1}, 99%, Euriotop), and incubated on deck for 24 h in the circulating incubators wrapped with neutral density screens to simulate *in-situ* irradiance conditions."

20. P7, L173-174: Before to compare the AOU data from the GEOVIDE program, you should explain how you get it. In facts, the section 2.7 is disturbing. There are two things here: the AOU and the comparison with historical data but there is no link between them. I suggest to split this section in two (even short) sections.

As suggested, we have split the original section 2.7 into two sections: section 2.7 Historical values and section 2.8 Apparent oxygen utilization as the following:

"**2.7 Historical values**

The historical data of the particulate ^{210}Po and ^{210}Pb activity, and the hydrological parameters (pressure, temperature, salinity, and dissolved oxygen) were obtained from databases and publications. The location, date, database address or publication name, and type of data (particulate ^{210}Po and ^{210}Pb activity or hydrological parameters) from all other studies is listed in supplemental Table S1.

2.8 Apparent oxygen utilization

Apparent oxygen utilization ($AOU = O_{2\ saturated} - O_{2\ measured}$) is defined as the difference between the saturated oxygen at a given temperature and salinity and the measured in-situ oxygen concentration (Ito et al., 2004; Duteil et al., 2013). A positive AOU indicates either water mass aging and outgassing of oxygen or biological activity, namely respiration (e.g. Keeling et al., 1998; Boyer et al., 1999). Negative AOU, indicating that the water is oversaturated with

dissolved oxygen, can appear under the conditions of an intense bloom (e.g. Coppola et al., 2017).

The dissolved oxygen concentration was measured by Winkler titration and the saturated oxygen concentration was calculated as a function of in-situ temperature and salinity, and one atmosphere of total pressure based on the built-in function in Ocean Data View (https://odv.awi.de)."

21. P2, L182-187: SPM, PP, chlorophyll were not considered to try to explain the 201Po-210Pb activities and activity ratios distribution?

The time-series chlorophyll-a data was considered to explain the distribution of total particulate $^{210}Po/^{210}Pb$ ratios < 1 at variable depths on L299-309 in the original manuscript.

The SPM and PP data, were indeed not used to try to explain the distribution of the radionuclide activities nor activity ratios. Instead, SPM were used to calculate the partitioning coefficient (K_d) while the in-situ PP and in-situ pigment data were considered to investigate the role of small particles in primary production and phytoplankton composition.

Results

22. p7, L195-202: there are a clear difference between station 1, 13, 21 and the other ones. These differences also correspond to the two samples groups that were processed by two labs. This is embarrassing if there is nothing that certify that labs results can be compared.

We agree there is a clear difference between stations 1, 13, 21 and the others along the transect. We acknowledge that the greater uncertainties of ^{210}Po and ^{210}Pb activities in the samples processed at QC were due to the longer delay between sampling and first plating (68 vs. 50 d) and higher uncertainties in the determination of the recovery of lead. It is unfortunate that we could not collect additional material to perform intercalibration between the two labs.

23. p7, L200-202: please rewrite this sentence which is very confusing.

We will change the sentence to: "$^{210}Po_t$ excesses relative to $^{210}Pb_t$, which were larger than $^{210}Po_t$ surface depletions at the same stations, were observed below the surface at some depths at stations 1, 13, and 21 in the Western European Basin (Fig. 2)."

24. p7, L195-207: this paragraph is confusing. Please describe firstly the surface water then the depth (or in the other way) but not a mixing description.

We concur. We will rewrite section 3.1 by describing first the activity range of all samples, then the surface samples, and last the deep samples. Section 3.1 will be changed to the following:

"Total ^{210}Po activities ($^{210}Po_t$) in all samples ranged from 2.2 to 16.4 dpm 100 L^{-1} and the mean $^{210}Po_t$ was 8.8 ± 2.4 dpm 100 L^{-1} (n = 198, Fig. 2). $^{210}Po_t$ activities were generally low within the mixed layer and euphotic zone (15 – 47 m), slightly increased or remained relatively constant in the depth range between the mixed layer and 250 m, and then decreased with water depth at most of the stations except station 26. Near the seafloor, stations 1, 13 and 44 had a slight increase of $^{210}Po_t$ activity.

Total ^{210}Pb activities ($^{210}Pb_t$) were between 2.1 and 20.6 dpm 100L^{-1} with a mean value of 10.0 ± 3.0 dpm 100 L^{-1} (n = 198, Fig. 2). $^{210}Pb_t$ activities were low in the surface, slightly increased in the subsurface and decreased with water depth. Stations 1, 13, 44, and 60 exhibited an increase near the seafloor.

The mean $^{210}Po_t/^{210}Pb_t$ activity ratio (AR) of all samples was 0.92 ± 0.28 (n = 198, Fig. 2). When considering different basins separately, there is a tendency of decreasing $^{210}Po_t/^{210}Pb_t$ AR from the Western European Basin (1.10 ± 0.35) westwards to the Iceland Basin (0.90 ± 0.19) and the Irminger Sea and the Labrador Sea (0.80 ± 0.18 and 0.83 ± 0.21, respectively).

For all regions, significant deficits of $^{210}Po_t$ (0.80 ± 0.20, n = 40) were observed within the mixed layer and euphotic zone (Fig. 3). Secular equilibrium was also observed at some shallow depths (i.e. 80 m at station 44) and even in surface waters (i.e. 15 m at station 38). $^{210}Po_t$ excesses relative to $^{210}Pb_t$, which were larger than $^{210}Po_t$ surface depletions at the same stations, were observed below the surface at some depths at stations 1, 13, and 21 in the Western European Basin (Fig. 2). At depths below the surface to ~ 1500 m in the Iceland Basin, the

Irminger Sea, and the Labrador Sea, the water samples still indicated a ^{210}Po deficiency (AR: 0.84 ± 0.17, n = 27). Secular equilibrium was generally reached near the bottom depths in all basins except at stations 13 and 60 where the water samples were either enriched in $^{210}Po_t$ ($^{210}Po_t/^{210}Pb_t$ AR = 1.58 ± 0.16) or depleted in $^{210}Po_t$ ($^{210}Po_t/^{210}Pb_t$ AR = 0.50 ± 0.12), respectively."

25. p8, L214-216: why the figure is not shown? The particulate profiles should be plotted (at least in the appendix material).

Thank you for the suggestion. The profiles of the particulate activity in the small and large size fractions will be shown as the following:

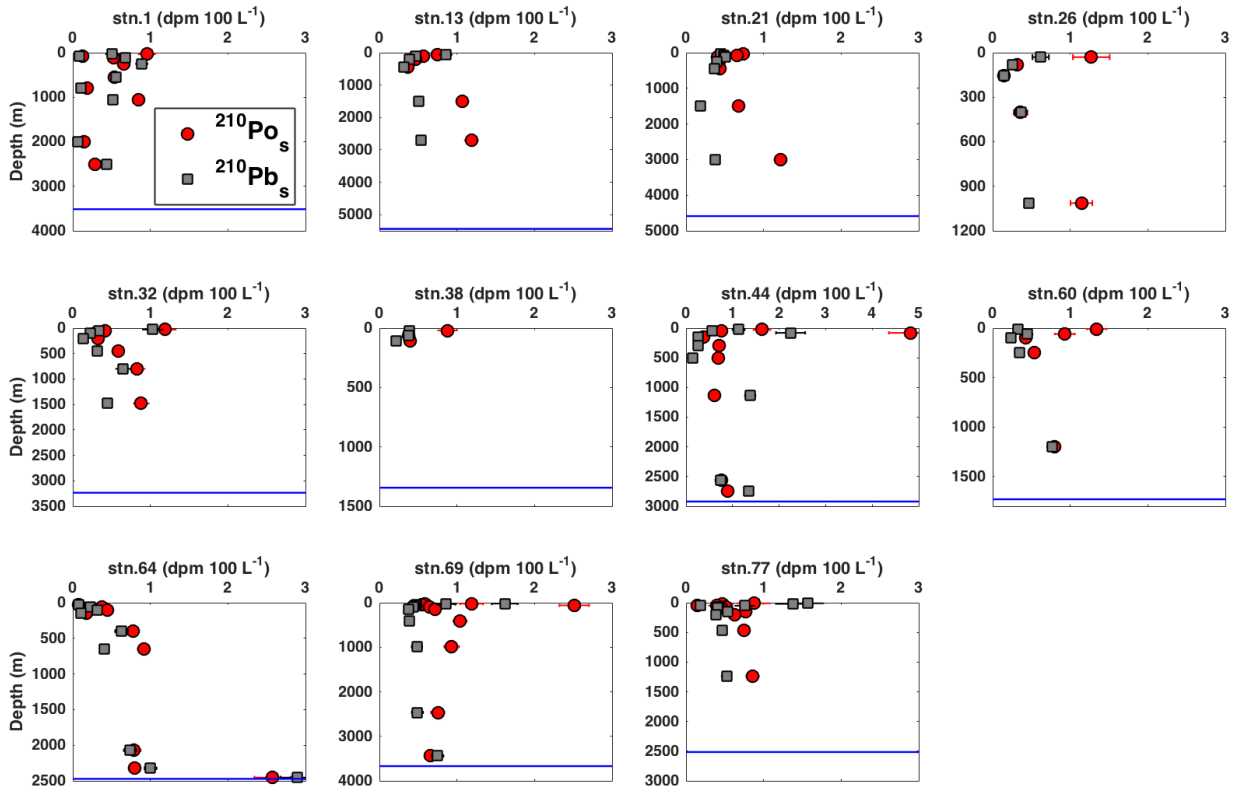


Fig. 4. Vertical profiles of particulate ^{210}Po and ^{210}Pb activity in the small size fraction (1-53 µm, $^{210}Po_s$, $^{210}Pb_s$). Note the different depth scales for the various stations and that the activity scale at Station 44 differs from the scale of all other stations. The horizontal blue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

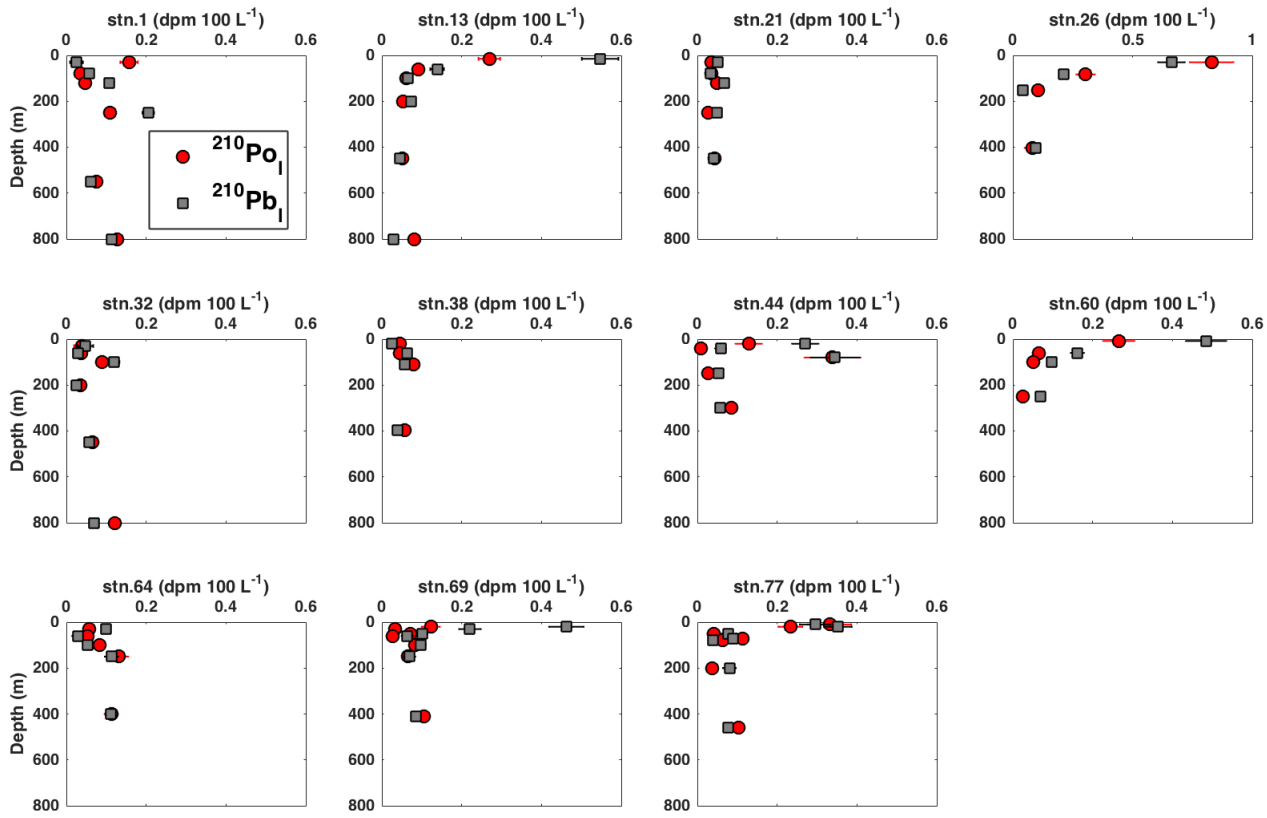


Fig. 5. The vertical profiles of the particulate ^{210}Po and ^{210}Pb activity in the large size fraction (> 53 µm, $^{210}Po_l$, $^{210}Pb_l$) in the top 800 m. Note that the activity scale at Station 26 differs from the scale at all other stations.

26. p9, L242-244: yes, this is not surprising as the small particle are the main particulate reservoir.

Yes, we agree. No change.

27. p9, L245-246: which particulate samples are depleted? Where they are located? In surface? Subsurface? Variable depths?

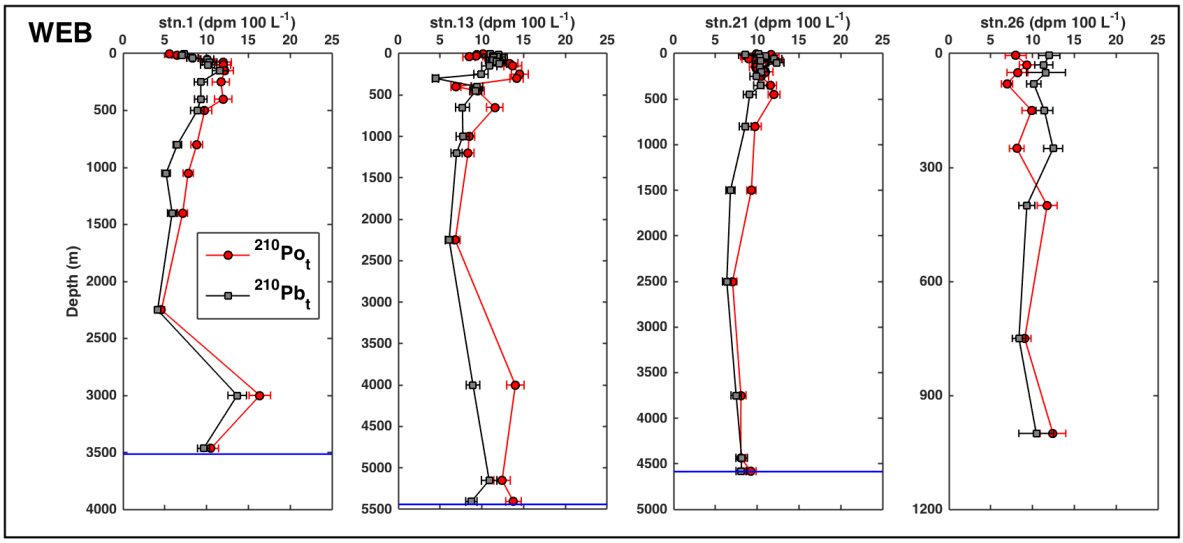
The information about those particulate samples with ^{210}Po/^{210}Pb AR < 1 was given in Table 1 and on Line 303-304. We will add the information here as: “While the majority of particulate

matter was enriched in ^{210}Po ($^{210}Po_p/^{210}Pb_p$ AR> 1), there were 13 out of 56 total samples from various depths that were depleted in ^{210}Po relative to ^{210}Pb."

Discussion

28. p10, L264-265: large excess is not seen at depth.

In the most recent submitted draft, we don't use the word "large", but we do see ^{210}Po activity excess at stations 1, 13 and 21 at depth. Please see the vertical profiles below (Fig. 2). In fact, the average $^{210}Po/^{210}Pb$ AR from 100 m down to the bottom depth was 1.2 ± 0.1, 1.4 ± 0.6, and 1.1 ± 0.1 at stations 1, 13, and 21, respectively.



29. p10, L260-267: I don't understand how an upwelling along the Iberian coast can bring excess 210Po all over the water column in the 3 station from the WEB.

We now rephrase the sentences on L260-267 as following:

"One possible source of these sub-surface ^{210}Po activity excesses below 200 m at stations 1 and 13 could be the North-East Atlantic Deep Water, lower ($NEADW_L$) which was the dominant water mass in the Iberian Basin from 2000 m to the bottom, and had a concentration of silicate up to 48 µmol kg^{-1} (García-Ibáñez et al., 2015). High activity of ^{210}Po in deep samples could be due to the dissolution of diatoms or herbivore feces (Cooper, 1952). As these particles sink and dissolve, ^{210}Po activity may have been preferentially released to the dissolved phase compared

to ^{210}Pb activity (Bacon et al., 1976), leading to ^{210}Po excess observed in the deep waters at stations 1 and 13. For the sub-surface ^{210}Po activity excesses at station 1 between 400 and 1000 m where lateral inputs of particulate Fe from the margin was observed (Gourain et al., 2018), the likely process is diffusion of ^{210}Po from those particles originated from the margin and such excess could be transported westwards to station 13 by lateral advection. An alternative source of ^{210}Po activity excess between 50 and 250 m at stations 1 and 13 (Fig. 3) could be the eastern boundary upwelling along the coast of the Iberian Peninsula (García-Ibáñez et al., 2015). Even though no strong upwelling events were revealed from temperature and density profiles during the cruise, northerly winds favoring upwelling were recorded 2 – 3 months before the sampling (Shelley et al., 2017). The deep water may have excess ^{210}Po activity due to the remineralization of sinking particles. The upwelling of this water mass prior to the sampling date could maintain such sub-surface excess ^{210}Po activity. Similar findings have been reported in the Cariaco Trench for the upper 300 m of the water column by Bacon et al. (1980)."

30. p11, L295-298: what do you mean by significant? Are they significantly different than this other station? Statistically tested? Is this confirmed from the data on the geochemical composition of SPM?

They are different from the other stations, but no statistical test was performed. Therefore, we will rephrase this sentence in the text as "In addition, the AR < 1 observed at station 1 (120, 250, and 550 m) could be associated with lithogenic particles from the Iberian Margin where 100% of the particulate Fe (PFe) had a lithogenic origin while the lithogenic contribution to PFe at other stations was smaller (Gourain et al., 2018)."

31. p11, L304-308: this is an interesting point. Is there a figure (or a way) to illustrate this? For example a plot showing the AR in surface or subsurface as a function of the time since the last bloom?

We appreciate the suggestions. We have plotted the depths at which total particulate ^{210}Po/^{210}Pb AR was found to be lower than unity as a function of the time since the last bloom in the following figure:

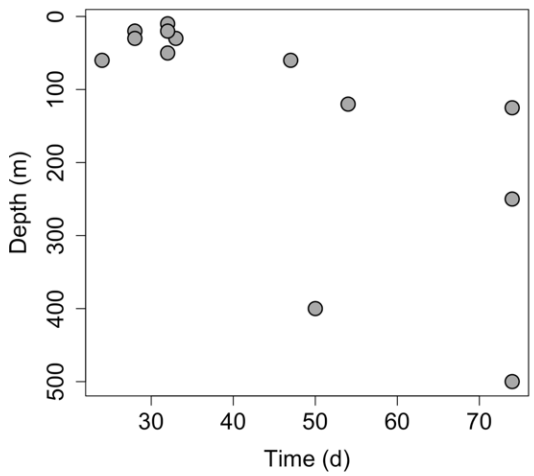


Fig. 7. Depths at which the total particulate (> 1 µm) ^{210}Po/^{210}Pb activity ratio was lower than unity vs. the time since the last bloom (data is presented in Table 1).

32. p12, L321: Is this particulate 210Po depletion in the coastal sea related to the 210Po/210Pb AR in these the terrestrial/riverine particles or is this due to the nature of those particles that present a lower scavenging efficiency of dissolved 210Po with respect to 201Pb?

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).

33. p12, L331: AOU must be defined in the method section. What a negative AOU value means?

Thank you for your suggestion. AOU is now defined in the method section 2.8 where the meaning of positive and negative AOU values are explained. Please see our response to specific comment 20.

34. p12, L332: remineralization + respiration + oxidation reactions.

We have corrected it. Please see that in the following response.

35. p12, L333-334: I do not see why water mass aging may change the OAU if there is no mineralization. To my opinion, only biogeochemical processes may change OAU values while the time can only change the intensity of O2 consumption by those biogeochemical processes. I think this should be better specified in this part to avoid confusion.

We agree that this sentence is not clear and will rephrase it as the following:

"AOU is a time-integrated measure of the amount of oxygen removed during the biogeochemical processes (e.g. respiration, remineralization, oxidation) in the ocean interior. Therefore, AOU is a product of apparent oxygen utilization rate (AOUR) and the age of water mass (e.g. Stanley et al., 2012), i.e. high AOU could be due to either intense biogeochemical processes that have occurred in a short period of time (young water mass) or weaker processes over a longer period of time (old water mass). Consequently, the rate of these biogeochemical processes and time (water mass age) would have different/similar impacts on the $^{210}Po_p/^{210}Pb_p$ AR value depending on the initial AR in the particles and the natural of the particles."

36. p12, L336: what is an old particle? Weeks? Months? Years?

It would be months to years as after 5 half-lives of ^{210}Po (~ 700 days), activity of ^{210}Po would be 95% of the activity of ^{210}Pb if there is no additional removal or addition of either isotopes.

37. p12, L336-338: time will induce an AR approaching 1: decreasing AR if the initial AR is >1 and increasing if the initial AR is < 1. Here you hypothesis that the initial AR in particle is <1 but both cases are possible. Please correct.

We agree and will change this sentence to "For example, the $^{210}Po_p/^{210}Pb_p$ AR would tend to increase with time if the initial AR is < 1 because particulate ^{210}Po activity would increase from the decay of ^{210}Pb and trend towards secular equilibrium ($^{210}Po_p/^{210}Pb_p$ AR = 1), and to decrease

with time if the initial AR is > 1 as the original excess of particulate ^{210}Po activity would disappear after 7 half-lives of ^{210}Po."

38. p12, L343-357: very interesting results and interpretation! However, I have two mains questions:

Thank you for your comment. Please see our responses to the two mains questions below.

39. Why the increase of AR from negative value to value close to 1 for OAU > 25 µmol/kg? Higher the OAU, higher the mineralization. So intuitively, the AR should be maintained more and more negative with increasing OAU?

We will rephrase the sentences on L350-355 as follows:

"The two contradictory linear trends likely reflect the nature of the particles. For example, the observation of $^{210}Po_p/^{210}Pb_p$ AR > 1 with AOU < 25 µmol kg^{-1} may suggest relatively fresh/organic particles in the young water mass. When AOU increases either due to water mass aging or higher AOUR, the $^{210}Po_p/^{210}Pb_p$ AR decreases with a slope of -0.17 ± 0.04. On the other hand, refractory/lithogenic particles may be suggested by the observation of $^{210}Po_p/^{210}Pb_p$ AR < 1 with AOU > 25 µmol kg^{-1}. For those particles, increasing in AOU either due to water mass aging or higher AOUR would change the $^{210}Po_p/^{210}Pb_p$ AR to a much lesser degree than that for organic particles with a slope of 0.008 ± 0.003."

Increasing AOU doesn't necessarily cause the AR to decrease if the particles are lithogenic or refractory.

40. I do not understand why it is said that this observation stands only for high latitude in the northern hemisphere. Other campaigns from high latitude in the Northern hemisphere are also reported on figure 5 but are not considered. In addition, GA-03 campaign are not from high latitude. What gives this relationship for other campaigns? Why this 4 campaigns was selected?

We found a general trend of lower particulate ^{210}Po/^{210}Pb AR in samples from relatively high latitude stations in the Northern Hemisphere. We wanted to specifically study the relationship between AR and AOU for those stations. However, we couldn't obtain the hydro-data

(temperature, salinity, dissolved oxygen) from the other campaigns at the high latitude in the Northern Hemisphere, therefore AOU could not be derived as it is a function of these parameters. That is why the other campaigns from the high latitude in the Northern Hemisphere reported on Figure 5 were not considered in the AR and AOU relationship on Figure 6.

We agree that GA03 is not from high latitude. But in the original manuscript when we included GA03 into the 4 campaigns, we obtained the two-phase correlation between AR and AOU on Figure 6 (AR< 1 & AOU > 25, AR> 1 & AOU < 25). It needs to be mentioned that the two-phase correlation still exists without GA03 data but R^2 will decrease to ~0.3 for both relationships with less data points. We investigated the ANT-X/6 in the Southern Ocean (AOU data only available at 13 stations) as it also seemed to have relatively low AR but the similar two-phase correlation between AR and AOU did not exist.

41. p13, L370: What do you mean by investigation of pigments? There is nothing about it in the material and methods section.

Yes, we indeed didn't include pigments in the methods section because the data has not been published yet.

We now removed the pigment data from the manuscript.

42. p14, L377-378: what do you mean by "as the above cited papers have seen elsewhere"? Please Precise

We now removed pigment and primary production from the manuscript. Please see our response to specific comment 47.

43. p14, L378-380: this is expected for the eastern part of the transect only?

Yes, it is. But it has been removed now. Please see our response to specific comment 41.

44. p14, L391-392: how did you calculate the dissolved activity? This is not indicated. When you consider the Kd for the small particles you normalize with the SPM for the small particles also? Same question for the total particulate. Please precise.

We calculated the dissolved activity by subtracting particulate activity from the total activity.

Yes, we used Eq. (1) to calculate K_d for both small and total particulate fractions, and K_d for small and total particulate fractions was normalized by the SPM in the small and particulate fractions, respectively.

We will clarify this as following:

"In this study, the size-fractionated data of radionuclide activity and SPM allowed us to calculate the partitioning coefficients for both radionuclides on small and total particles by using Eq. (1). The dissolved radionuclide activity was calculated as the difference between total and particulate activity. The coefficients for the small and total particulate phases were normalized by the SPM in the small and total particulate phases, respectively."

45. p14, L399-401: How this is possible as the small particulate activity is necessary lower than the total particulate activity? Is it associated to the SPM normalization?

Yes, it is indeed associated to the SPM normalization. Please see our previous response.

46. p14, L401-403: here you affirm that the scavenging and export is mostly driven by small particles. But there is nothing to confirm this. Although this can be plausible, this is just an hypothesis.

We agree that we don't have direct evidence of small particles sinking nor that export was driven by the small particles. We therefore removed this topic from this manuscript and will discuss it further in the companion paper. However, the comparison of K_d for both radionuclides in the small size fraction vs. total size fraction suggests that the adsorption/scavenging of radionuclides were driven by the small particles.

47. p12-14, L362-404: this section is very surprising. From the title of the section I excepted to find POC export calculation. In facts, there is no data really discussed or even showed

(pigment, primary production, …) and most of the discussion is based on hypothesis without real solid basis to support them. I suggest to rewrite this section around concrete data only and to change the title of this section.

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.

We agree that the title of this section is not appropriate. We will change it to "Relationship among small particles, adsorption, and scavenging".

We now removed the statements about the pigment and primary production from this section and will rewrite it as follows:

"4.4 Relationship among small particles, adsorption, and scavenging

The partitioning coefficient, K_d (L kg^{-1}), has been used to describe the particle adsorption behavior of radionuclides. It is defined as the ratio of the adsorbed radionuclide activity (A_p, dpm 100L^{-1}) to the dissolved radionuclide activity (A_d, dpm 100L^{-1}), normalized by the suspended particulate matter concentration (SPM, µg L^{-1}):

$$K_d = \frac{A_p}{A_d} \times \frac{1}{SPM} 10^9 \qquad (1)$$

Owing to the different biological and chemical behaviors of ^{210}Po and ^{210}Pb, the interpretation of measured K_d for ^{210}Po (K_d(Po)) may not be as clear as that for ^{210}Pb (K_d(Pb)). 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.

In this study, the size-fractionated data of both radionuclide activity and SPM allowed us to calculate the partitioning coefficients for both radionuclides on small and total particles. The dissolved radionuclide activity was calculated as the difference between total and particulate activity. The coefficients for the small particulate and the total particulate phases were

normalized by the SPM in the small and total particulate phases, respectively. We present only the coefficients for the small particulate phases ($K_d(Po)_s$, $K_d(Pb)_s$) and the total particulate phases ($K_d(Po)_p$, $K_d(Pb)_p$) because most of the particulate activity (> 80%) was associated with the small particles along the GEOVIDE transect, and most conceptualized scavenging models consider either the two-box model (dissolved – total particulate phases, i.e. $K_d(Po)_p$) or the three-box model (dissolved – small – large, i.e. $K_d(Po)_s$) (Clegg and Whitfield, 1990; 1991; Rigaud et al., 2015) and thus activity is concentrated from the dissolved phase to the total or small particles.

The average values of $K_d(Po)$ was 1.6 times of those of $K_d(Pb)$ in both small and total particulate phases, suggesting a higher affinity with particles for ^{210}Po with respect to ^{210}Pb, which is commonly observed in the global ocean (Bacon et al., 1988; Hong et al., 1999; Masqué et al., 2002; Wei et al., 2014; Tang et al., 2017). The K_d values for the small particulate phase were slightly higher than those for the total particulate phase but overall these values were very similar for both radionuclides (Fig. 7), suggesting that adsorption/scavenging of radionuclides was driven by small particles along the transect. In addition, there are increasing studies which argue that small particles can form aggregates that sink, and their contribution to carbon export could be larger than previously thought (e.g. Richardson and Jackson, 2007; Lomas and Moran, 2011; Amacher et al., 2013; Puigcorbé et al., 2015). We, therefore, recommend combining the activities of both small and large particles into a total particulate fraction in order to explain total $^{210}Po/^{210}Pb$ disequilibria in the surface waters, and utilizing the characteristics of the total particles (instead of just the large particles) in the estimation of the POC export fluxes (Tang et al., companion paper submitted to this volume).

Traditionally, large particles collected by in-situ filtration with pumps, most commonly defined as particles larger than 53 or 70 µm, were assumed to dominate the sinking flux (Dugdale and Goering, 1967; Bishop et al., 1977; Fowler and Knauer, 1986; Honjo et al., 1992; Walsh and Gardner, 1992) such that the composition ($POC/^{210}Po$) of the large particle size class was used to convert ^{210}Po fluxes into POC export (e.g. Friedrich and Rutgers van der Loeff, 2002; Cochran and Masqué, 2003; Murray et al., 2005; Stewart et al., 2010; Roca-Martí et al., 2016). Given that the true size spectrum of sinking particles for the timescale relevant to the

$^{210}Po/^{210}Pb$ method is unknown and the POC flux estimates are sensitive to the particulate POC/^{210}Po ratio, both small and large particles should be sampled for POC/^{210}Po due to the variability in the POC/^{210}Po ratio in different size classes (Hayes et al., in review)."

Conclusion:

48. p15, L415-420: again this was not clearly demonstrated. This conclusion should be very robust because it can have large implications in the future sampling strategy. Differently: does the sampling and analysis of two particulate size fractions is necessary in the future? So this has to be very robustly demonstrated. I agree with the fact that the high proportion of particulate nuclides is found in the small particle indicates that small particles are important in the sorption process. But I'm clearly not convinced from the data showed in the manuscript there is evidence to say that the small particles play an important role in the export of particles. If so, this should be strengthened. I may suggest to synthesis the most important findings based on the data only. There is nothing on the time elapsed since the last bloom for example.

We agree that conclusions should be based on the data only and, therefore, we will remove the text about the relationship between small phytoplankton and export. Conclusions will be changed to the following:

"In this study, we reported the vertical distribution of total and size-fractionated particulate ^{210}Po and ^{210}Pb activities in the North Atlantic during the GEOVIDE GA01 cruise. More than 90% of the radionuclide activity was found in the dissolved phase, while a small proportion was associated with particles in this transect. Total ^{210}Po activity was generally depleted relative to total ^{210}Pb activity in the upper 100 m due to the preferential adsorption of ^{210}Po activity by particles. Such deficiencies of ^{210}Po activities generally extended to the deep waters at most of the stations. In the Western European Basin, the excess of ^{210}Po activities at stations 1 and 13 in the North East Atlantic Deep Water was attributed to the release of ^{210}Po during dissolution of sinking biogenic particles.

There appear to be geographic differences in particulate $^{210}Po/^{210}Pb$ activity ratios measured during GEOVIDE and previous studies, with particularly low values in the high-latitude North Atlantic and Arctic. While this observation deserves more attention, we support previous suggestions that this is due to the terrestrial origin/riverine input of particles with a low $^{210}Po/^{210}Pb$ AR into the river-dominated shallow seas of the Arctic. The age of the particles and water masses as well as the importance of biogeochemical processes (e.g. respiration, remineralization) may also explain some of these observations, as there was a significant relationship between the total particulate activity ratio and AOU when both were measured in the North Atlantic (> 20 ºN) and Arctic Oceans.

Over 80% of the particulate radionuclide activity was on small particles, indicating that the scavenging of both radionuclides was driven by small particles. Therefore, we suggest considering the activities of ^{210}Po and ^{210}Pb from both small and large particles in order to study the water column $^{210}Po/^{210}Pb$ disequilibria and quantify POC export along the GA01 transect. This has been addressed in a companion paper in this issue. We recommend that both small and large particles should be sampled for $POC/^{210}Po$ estimates for the application of the $^{210}Po/^{210}Pb$ method in future studies of POC export."

Fig 3:

49. I doubt the sentence "A closer look at only the zoom" is correct in the caption

Those words will be removed from the caption. The caption will be changed as: "The upper 250 m of the depth profiles of total ^{210}Po ($^{210}Po_t$, red circles) and ^{210}Pb activities ($^{210}Pb_t$, grey squares) along the GEOVIDE section. The horizontal orange and magenta lines denote the mixed layer depth (MLD) and the base of the euphotic zone ($Z_{1\%}$), respectively. The depth profiles are shown in the order of sampling and grouped by region (refer to Fig. 2 for the text abbreviations)."

50. Stn 60: 2 dot are missing for 210Pb at approximatively 50 m and 120 m depth.

The range of horizontal axis (activity) for each plot will be changed from 0-20 to 0-25 dpm 100 L^{-1} and the two data points will be shown in the plot of stn. 60. Please see the plot below:

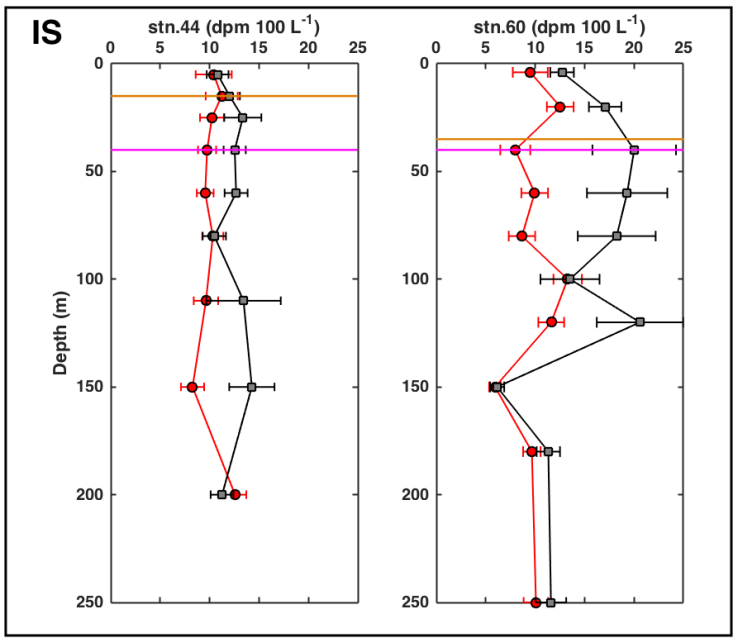


Figure 6:

51. negative AOU value need to be explained?

Negative AOU value is now explained in the text. Please see our response to specific comment 20.

52. with the uncertainty on Po/Pb AR there is (most of the time) not significant deviation from the 1 AR for the "other points". I suggest to integrate the "other points" within the regression keeping the only separation lower or above 25 µmol/kg for OAU.

We agree. The original Figure 6 will be changed to the following:

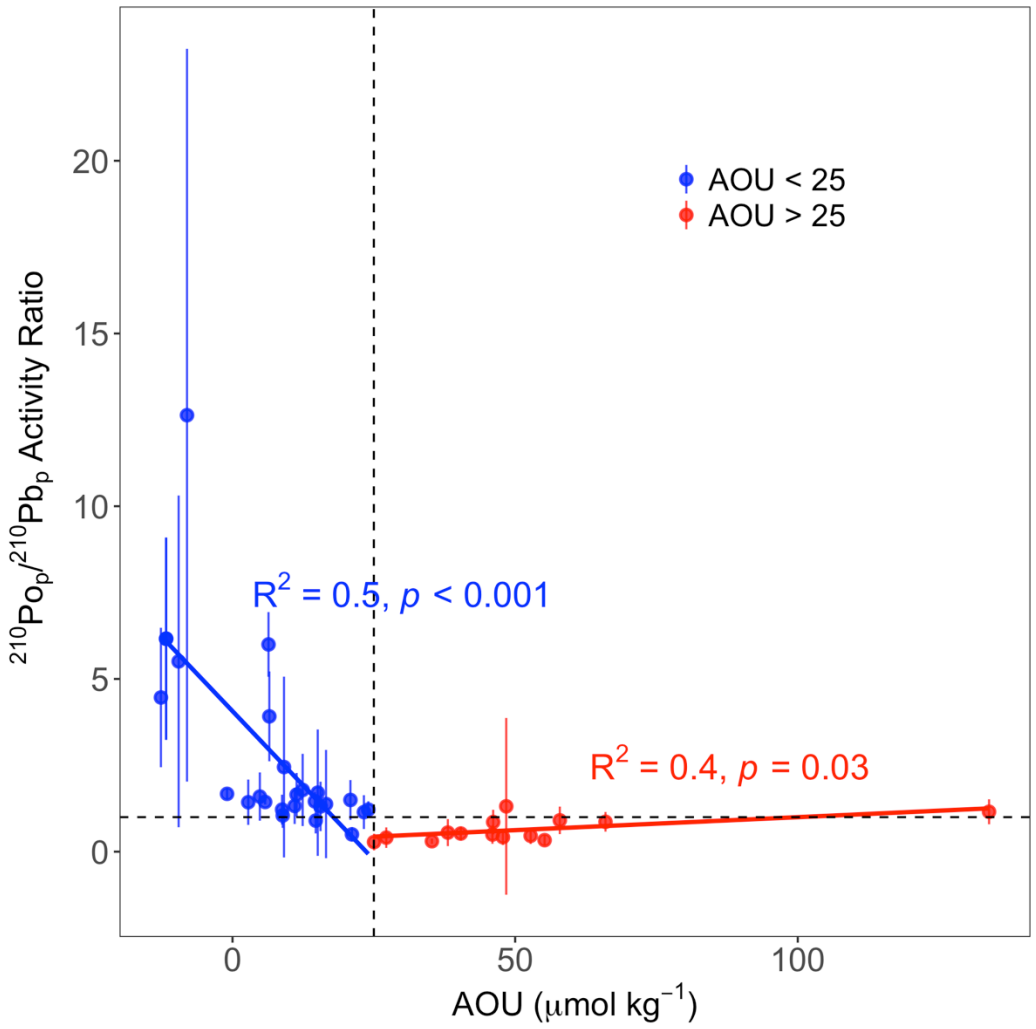


Fig. 9. The relationship between AOU (µmol kg^{-1}) and total particulate ^{210}Po/^{210}Pb activity ratio ($^{210}Po_p/^{210}Pb_p$) from the upper 200 m in the northern hemisphere (> 22 ºN) investigated by a linear regression model (red and blue lines). The 40 stations include data from previous studies, ARK-XXII/2 (77.38-87.83 ºN, n = 15) in the Arctic, BOFS (48.89-49.87 ºN, n = 7), GA03 (22.38-39.70 ºN, n = 7), and this study, GA01 (40.33-59.80 ºN, n = 11) in the North Atlantic. The horizontal dashed line represents $^{210}Po_p/^{210}Pb_p$ AR = 1 and the vertical dashed line represents AOU = 25 µmol kg^{-1}. Blue circles denote AOU < 25 µmol kg^{-1}, while red circles denote AOU > 25 µmol kg^{-1}.

Figure 7:

53. the axis labels on the figure and in the caption are not the same. Please homegeneize.

We agree. The caption of Figure 10 (no longer 7 because of the addition of figures) will be changed as:

"Comparison of the partitioning coefficient (K_d) between the dissolved and small particulate phases ($K_d(Po)_s$, $K_d(Pb)_s$) vs. between the dissolved and total particulate phases ($K_d(Po)_p$, $K_d(Pb)_p$) for (a) ^{210}Po and (b) ^{210}Pb. The 1:1 line is indicated as the solid line in each plot."

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Distributions of total and size-fractionated particulate ^{210}Po and ^{210}Pb activities along the North Atlantic GEOTRACES GA01 (GEOVIDE) ~~cruise~~ transect~~: partitioning between the particulate and dissolved phase~~

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Abstract

Vertical distributions of total and particulate ^{210}Po and ^{210}Pb activities in the water column were measured at eleven stations in the North Atlantic during the GEOTRACES GA01 GEOVIDE cruise in May - June 2014. Total ^{210}Po activity was on average 24% lower than ^{210}Pb activity in the upper 100 m, and was closer to unity in the mesopelagic (100 – 1000 m). The partitioning coefficients (K_d) along the transect suggest the preferential association of ^{210}Po relative to ^{210}Pb onto particles. The prominent role of small particles in sorption was confirmed by the observation that over 80% of the particulate radionuclide activity was on small particles. To account for the observed surface water ^{210}Po/^{210}Pb disequilibria, particulate radionuclide activities and export of both small (1-53 μm) and large (> 53 μm) particles must be considered. A comparison between the GEOVIDE total particulate ^{210}Po/^{210}Pb activity ratios (AR) and the ratios in previous studies revealed a distinct geographic distribution, with lower particulate AR in the high-latitude North Atlantic (including this study) and Arctic in relation to all other samples. For the samples where apparent oxygen utilization (AOU) was calculated at the same depth and time as the ^{210}Po/^{210}Pb AR (40 stations including this study), there was a two-phase correlation between the total particulate AR and AOU likely reflecting the nature of the particles and demonstrating the ~~competing~~ forces of remineralization and radionuclide decay from particles as they age.

1 Introduction

The major goal of the international GEOTRACES program is to characterize the distributions of trace elements and isotopes (TEIs) in the ocean on a global scale, and to identify and quantify processes that control these distributions (GEOTRACES Planning Group, 2006). The GEOVIDE section was a contribution of the French GEOTRACES program to this global program in the subpolar North Atlantic. The GEOVIDE GA01 cruise was carried out in 2014 in the North Atlantic and consisted of two sections: a section along the OVIDE (Observatoire de la variabilité interannuelle et décennale en Atlantique Nord) line between Lisbon (Portugal) and Cape Farewell (southern tip of Greenland), and a Cape Farewell to St. John's (Canada) section across the Labrador Sea (Fig. 1). Since 2002, the OVIDE section has been occupied biennially to collect physical and biogeochemical data (Mercier et al., 2015). The knowledge of the currents, water masses, and biogeochemical provinces gained from the previous OVIDE campaigns enabled the optimal strategy for TEIs sampling and provided help for the interpretation of the distribution of TEIs in the subpolar North Atlantic (García-Ibáñez et al., 2015). In addition to the OVIDE line, the Labrador Sea section provided a unique opportunity to study TEIs distributions along the boundary current of the western North Atlantic subpolar gyre (Sarthou et al., in review). ~~The major goal of the international GEOTRACES program is to characterize the distributions of trace elements and isotopes (TEIs) in the ocean on a global scale, and to identify and quantify processes that control these distributions (GEOTRACES Planning Group, 2006). The GEOVIDE section was a contribution of the French GEOTRACES program to this global survey in the North Atlantic. The GEOVIDE GA01 cruise was carried out in 2014 in the North Atlantic at latitudes greater than 40 ºN and consisted of two sections: the seventh repetition of the OVIDE section from Lisbon (Portugal) to Cape Farewell (southeast tip of Greenland), and a Cape Farewell to St. John's (Canada) section across the Labrador Sea (Fig. 1). The water mass properties and main current transports have been well studied in the OVIDE section during six previous repeated hydrological surveys (2002-2012) (García-Ibáñez et al., 2015). Conditions along the Cape Farewell-St. John's section, however, were relatively unknown. The combination of the two sections constitutes a mixture of complex water masses, circulation patterns, and oceanic boundaries, presenting a special opportunity to analyze the rates of the processes that govern the distribution of TEIs.~~

Polonium-210 (^{210}Po, $T_{1/2}$ = 138.4 d) and its radioactive grandparent Lead-210 (^{210}Pb, $T_{1/2}$ = 22.3 y) are two non-conservative ^{238}U decay series products. The GEOTRACES program has

included both radionuclides in its TEIs list primarily due to ^{210}Po's enhanced bioaccumulation and the use of the ^{210}Po/^{210}Pb pair as a proxy for assessing particle export in the upper ocean. 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; Ceballos-Romero et al., 2016), 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). However, since the data reported by Bacon et al. (1980b) at the Labrador Sea stations (47.8 – 53.7 °N), there are few studies of ^{210}Po and ^{210}Pb activity in the North Atlantic at latitudes greater than 40 °N. The GEOVIDE cruise, which targeted the North Atlantic from 40 °N to 60 °N, provided an opportunity to fill this data gap.

Besides ascertaining the distribution of the natural radionuclides under specific geographic conditions, this project aimed to answer questions about their biogeochemical behaviors in various marine environments. Owing to the significantly longer half-life of ^{210}Pb relative to ^{210}Po, the two radionuclides are expected to be in secular equilibrium (total ^{210}Po/^{210}Pb activity ratio = 1) in the ocean, assuming no net removal or addition of either radionuclide. A deficit of ^{210}Po activity relative to ^{210}Pb activity (^{210}Po/^{210}Pb activity ratio < 1), however, is commonly found in the upper ocean (e.g. Bacon et al., 1976; Nozaki and Tsunogai, 1976; Cochran et al., 1983; Sarin et al., 1999). This has been attributed to a higher particle reactivity of ^{210}Po (higher partitioning coefficient, K_d) than ^{210}Pb in seawater. Particles, therefore, become enriched in ^{210}Po (^{210}Po/^{210}Pb activity ratio > 1) and their sinking to deeper waters results in a ^{210}Po activity deficit relative to ^{210}Pb activity in the upper water column where particles are formed.

In this work, we describe the distributions of total and size-fractionated particulate ^{210}Po and ^{210}Pb activity along the GEOVIDE cruise in the North Atlantic. These data are a significant contribution to the high-latitude North Atlantic ^{210}Po and ^{210}Pb activity data set. We present a compilation of particulate ^{210}Po/^{210}Pb activity ratios (AR) from previous studies in the global ocean and the results are discussed in regards to the aging of water and biochemical processes. We also describe the relationship among small particles, adsorption, and scavenging of radionuclides. These results lead to recommendations for the estimation of particulate organic carbon export flux based on the ^{210}Po/^{210}Pb disequilibrium, a topic that is covered in a companion paper (Tang et al., , companion paper submitted to this volume).~~In this work, we present the distributions of total and~~

~~particulate ^{210}Po and ^{210}Pb activity at 11 stations along the GEOVIDE cruise. These data are a significant contribution to the high-latitude North Atlantic ^{210}Po and ^{210}Pb activity data set. In addition, we calculate the K_d of ^{210}Po and ^{210}Pb during scavenging, discuss why this value has a complicated interpretation, and is mostly likely driven by sorption to small particles. We also put our somewhat unusually low particulate ^{210}Po/^{210}Pb activity ratios (AR) into a global context and look for any possible cause of variation along the cruise path.~~

2 Methods

2.1 Sample collection

The French GEOTRACES cruise to the North Atlantic (GEOVIDE, Section GA01; May 15 – June 30, 2014) was completed on the ~~*N/O*~~ *R/V Pourquoi Pas?*. The research vessel departed from Lisbon, Portugal, headed northwest to the Greenland shelf, crossed the Labrador Sea, and ended in St John's, Newfoundland, Canada (Fig. 1). A rosette equipped with conductivity-temperature-depth sensors and 12 L Niskin bottles was used to collect 200 seawater samples (5 – 10 L each) from 10 full water column "super" (10 multi-cast) stations (16 – 22 depths/station) and 1 "~~Xlarge~~ XLarge" (5-cast) station to 800 m (station 26, 9 depths) for the determination of total ^{210}Po and ^{210}Pb activity. Upon recovery, seawater samples were transferred to 10 L acid-cleaned containers. In addition, particulate radionuclide activities in two size classes (1-53 µm and > 53 µm) were collected at 3 – 10 depths per station using large volume *in-situ* filtration systems (Challenger Oceanic pumps and McLane pumps) equipped with 142 mm filter holders. Each filter head contained a stacked 53 µm PETEX screen followed by a 1 µm pore size quartz fiber QMA filter. The volume filtered was determined via flow meters mounted below each filter head, and the mean volume pumped through each head was 881 L. Once recovered, clear polyethylene caps were placed on the top of the pump heads and they were brought into a clean laboratory for sub-sampling.

2.2 Total ^{210}Po and ^{210}Pb

Total ^{210}Po and ^{210}Pb activities were determined from the seawater samples by the cobalt-ammonium pyrrolidine dithiocarbamate (Co-APDC) technique (Fleer and Bacon, 1984). Samples were acidified to a pH < 2 with concentrated HCl immediately after collection and spiked with known amounts of ^{209}Po and stable lead as chemical yield tracers. After vigorous stirring and at least ~~6~~ 12 h of isotope equilibration, cobalt nitrate and APDC solutions were added to co-

precipitate Po and Pb. Samples were filtered through a 0.45 µm membrane filter and the filters with the precipitate were placed ~~transferred~~ into ~~a~~ clean falcon tubes ~~bottle~~, sealed with parafilm, and stored in double-bags. As the delay between sample collection and first Po plating increases, the uncertainty of the calculated ^{210}Po activity also increases. In addition, it is necessary to balance counting periods with the number of samples as the uncertainty due to alpha spectrometry counting decreases by increasing the counting time. To limit the delay between sampling and processing and to ensure higher counting statistics by having more alpha spectrometers devoted to this project, sample processing and analyses were split between Universitat Autònoma de Barcelona (UAB) (samples from stations 1, 13, and 21) and Queens College (QC) (stations 26, 32, 38, 44, 60, 69, and 77). ~~Further sample processing and analyses were split between the Laboratori de Radioactivitat Ambiental (LRA) at Universitat Autònoma de Barcelona (UAB) (samples from stations 1, 13, and 21) and the Stewart laboratory at Queens College (QC) (stations 26, 32, 38, 44, 60, 69, and 77) to ensure higher counting statistics in the samples.~~ Both laboratories followed the same procedure. Briefly, the filters were digested into a solution of concentrated HNO_3 and HCl, and after the solution was evaporated to dryness, the samples were recovered in 1M and 0.5 M HCl solution at UAB and QC, respectively (a 0.5-2 M HCl solution is recommended, Rigaud et al., 2013). ~~Briefly, the filters were digested in a mixture of concentrated HNO_3 and HCl, evaporated to dryness, and eventually dissolved in 1M and 0.5 M HCl at UAB and QC, respectively.~~ A polished pure silver disc (Flynn, 1968) with one side covered by enamel paint was placed into the weak acid solution and heated so that the polonium nuclides were spontaneously plated onto only one side of the disc. The activities of both Po nuclides on the disc were measured by alpha spectrometry. Any ^{210}Po and ^{209}Po remaining in the plating solution was removed using AG 1-X8 anion exchange resin and the final solution was re-spiked with ^{209}Po and stored for more than 6 months to allow ingrowth of ^{210}Po from the decay of ^{210}Pb.

The ^{210}Pb activity was then determined by re-plating the solutions using silver discs and measuring the ingrown ^{210}Po. Two aliquots of the plating solutions for each sample were taken before the first and second platings for the measurement of total Pb concentration by inductively coupled plasma mass spectrometry (ICP-MS) to determine sample recovery during processing. The average recoveries produced by ~~the LRA~~UAB and ~~Stewart groups~~ QC were 83 ± 11% (n = 54) and 76 ± 14% (n = 144), respectively. The activities of ^{210}Po and ^{210}Pb at the time of collection were determined by a series of corrections, including nuclide decay, ingrowth, chemical recoveries,

detector backgrounds, and blank contamination following the methods in Rigaud et al. (2013). The activity uncertainties from UAB were on average 8% for both ^{210}Po and ^{210}Pb activity, while the QC uncertainties were on average 13% for ^{210}Po activity and 16% for ^{210}Pb activity. The greater uncertainties of ^{210}Po and ^{210}Pb activities in the samples processed at QC were due to the longer delay between sampling and first plating (68 vs. 50 d) and higher uncertainties in the determination of the recovery of lead. ~~Finally, the initial activities of ^{210}Po and ^{210}Pb at the time of collection were determined by a series of corrections, including nuclide decay, ingrowth, chemical recoveries, detector backgrounds, and blank contamination following the methods in Rigaud et al. (2013). The activity uncertainties from LRA were on average 8% for both ^{210}Po and ^{210}Pb activity, while the activity uncertainties from the Stewart group were on average 13% for ^{210}Po activity and 16% for ^{210}Pb activity.~~

2.3 Particulate ^{210}Po and ^{210}Pb

After collection via in situ pumping, one quarter (equivalent to ~ 220 L) of the PETEX screen containing > 53 µm or "large" particles was processed for radionuclide activity. Swimmers were carefully removed from all samples. The QMA filters containing 1-53 µm or "small" particles were sub-sampled (2 – 4 punches of 12 mm-diameter) achieving a mean effective volume of ~ 66 L. The screens and punches were stored in double-bags at -80 °C until the analyses onshore. The particulate samples were split between the two laboratories in parallel to the seawater samples. The filters were spiked with ^{209}Po tracer solution and stable lead, digested using a mixture of concentrated HF, HNO_3 and HCl at UAB, but only HNO_3 and HCl at QC. After multiple rounds of digestion and evaporation to near dryness, the samples were recovered in 0.5 M HCl solution. Any remaining pieces of filter which were not completely digested were carefully removed, rinsed with 0.5 M HCl solution several times, and then discarded. The analyses of the particulate radionuclide activities were identical to those for the seawater samples described in section 2.2.

2.4 Concentration of suspended particulate matter (SPM)

The Helene Planquette group (University of Brest, co-authors in this issue) collected subsamples from the same screens and filters that were sampled previously for radionuclides to determine major phase composition (particulate organic matter (POM), lithogenic material, calcium carbonate ($CaCO_3$), opal, $Fe(OH)_3$, and MnO_2) (references therein Lam et al.,

2015)~~utilized the material on the balance of the screens and filters after subsampling for radionuclides to determine major phase composition (particulate organic matter (POM), lithogenic material, calcium carbonate ($CaCO_3$), opal, $Fe(OH)_3$, and MnO_2) (references therein Lam et al., 2015)~~. The ~~complete details of sampling and analyses will be described in a separate manuscript (Lemaitre et al., in prep.), but the~~ mass concentration of ~~total~~ SPM was calculated as the sum of the chemical dry weight of the major particulate phases.

The calculated SPM concentration was compared to the *in-situ* transmission data obtained from the rosette CTD sensor (Fig. S1). The overall negative relationship was statistically significant (R^2 = 0.7, n = 53, $p < 0.0001$), suggesting that the SPM concentrations determined were reasonable estimates of particle concentration in the water column. We used the SPM values to determine the partitioning coefficient, K_d, for ^{210}Po and ^{210}Pb in section 4.4.

~~**2.5 Primary production**~~

~~Daily primary production (PP) at each station was determined using the ^{13}C labeling technique by the Dehairs group. The details of sampling and analysis for PP is presented in depth elsewhere (Fonseca-Batista et al., 2018). Briefly, seawater samples (3 – 6 depths/station) were collected from the surface to the depth of 0.2% photosynthetically active radiation (PAR). The seawater was then incubated on deck for 24 h under conditions of photometric depths. After incubation, seawater was filtered through GF/F filters (0.7 µm porosity), followed by ^{13}C determination using elemental analysis-isotope ratio mass spectrometry. Daily PP was derived from the depth-integrated ^{13}C uptake rates.~~

2.5 Satellite-based data

The 8-day composites of surface chlorophyll-a concentration for each station were retrieved from NASA's MODIS products (https://oceancolor.gsfc.nasa.gov) for the period from January to July 2014. The time-series chlorophyll-a concentrations were used to show the development of a phytoplankton bloom over time along the transect.

2.6 ~~Apparent oxygen utilization and h~~ Historical values

The historical data of the particulate ^{210}Po and ^{210}Pb activity, and the hydrological parameters (pressure, temperature, salinity, and dissolved oxygen) were obtained from databases and

publications. The location, date, database address or publication name, and type of data (particulate ^{210}Po and ^{210}Pb activity or hydrological parameters) from all other studies is listed in supplemental Table S1.

~~We compared the GEOVIDE data (particulate radionuclide activity and apparent oxygen utilization) to historical databases and publications. The apparent oxygen utilization (AOU, µmol kg^{-1}), a measurement of respiration and water mass age (Stanley et al., 2012), can be derived from hydrological parameters (pressure, temperature, salinity, and dissolved oxygen) using the built-in function in Ocean Data View. The location, date, database address or publication name, and type of data (particulate ^{210}Po and ^{210}Pb activity or hydrological parameters) from all other studies is listed in the supplemental Table S1.~~

2.7 Apparent oxygen utilization

Apparent oxygen utilization ($AOU = O_{2\,saturated} - O_{2\,measured}$) is defined as the difference between the saturated oxygen at a given temperature and salinity and the measured in-situ oxygen concentration (Ito et al., 2004; Duteil et al., 2013). A positive AOU indicates either water mass aging and outgassing of oxygen or biological activity, namely respiration (e.g. Keeling et al., 1998; Boyer et al., 1999). Negative AOU, indicating that the water is oversaturated with dissolved oxygen, can appear under the conditions of an intense bloom (e.g. Coppola et al., 2017).

The dissolved oxygen concentration was measured by Winkler titration and the saturated oxygen concentration was calculated as a function of in-situ temperature and salinity, and one atmosphere of total pressure based on the built-in function in Ocean Data View (https://odv.awi.de).

2.8 Statistical analyses

Statistical analyses were carried out in R Studio version 3 using Fitting Linear Models, and Welch Two Sample t-tests. Linear regression analysis was used to investigate the relationship between total particulate ^{210}Po/^{210}Pb AR and AOU. The Welch Two Sample t-test was applied to assess whether the mean of the total particulate ^{210}Po/^{210}Pb AR was the same as the mean of the small particulate ^{210}Po/^{210}Pb AR. It was also applied to investigate the means of the total ^{210}Pb activity in the western and eastern sections along the transect.

3 Results

3.1 Total ^{210}Po and ^{210}Pb activities

Total ^{210}Po activities ($^{210}Po_t$) in all samples ranged from 2.2 to 16.4 dpm 100 L^{-1} and the mean $^{210}Po_t$ was 8.8 ± 2.4 dpm 100 L^{-1} (n = 198, Fig. 2). $^{210}Po_t$ activities were generally low within the mixed layer and euphotic zone (15 – 47 m), slightly increased or remained relatively constant in the depth range between the mixed layer and 250 m, and then decreased with water depth at most of the stations except station 26. Near the seafloor, stations 1, 13 and 44 had a slight increase of $^{210}Po_t$ activity.

Total ^{210}Pb activities ($^{210}Pb_t$) were between 2.1 and 20.6 dpm 100L^{-1} with a mean value of 10.0 ± 3.0 dpm 100 L^{-1} (n = 198, Fig. 2). $^{210}Pb_t$ activities were low in the surface, slightly increased in the subsurface and decreased with water depth. Stations 1, 13, 44, and 60 exhibited an increase near the seafloor.

The mean $^{210}Po_t/^{210}Pb_t$ activity ratio (AR) of all samples was 0.92 ± 0.28 (n = 198, Fig. 2). When considering different basins separately, there is a tendency of decreasing $^{210}Po_t/^{210}Pb_t$ AR from the West European Basin (1.10 ± 0.35) westwards to the Iceland Basin (0.90 ± 0.19) and the Irminger Sea and the Labrador Sea (0.80 ± 0.18 and 0.83 ± 0.21, respectively).

For all regions, significant deficits of $^{210}Po_t$ (0.80 ± 0.20, n = 40) were observed within the mixed layer and euphotic zone (Fig. 3). Secular equilibrium was also observed at some shallow depths (i.e. 80 m at station 44) and even in surface waters (i.e. 15 m at station 38). $^{210}Po_t$ excesses relative to $^{210}Pb_t$, which were larger than $^{210}Po_t$ surface depletions at the same stations, were observed below the surface at some depths at stations 1, 13, and 21 in the West European Basin (Fig. 2). At depths below the surface to ~ 1500 m in the Iceland Basin, the Irminger Sea, and the Labrador Sea, the water samples still indicated a ^{210}Po deficiency (AR: 0.84 ± 0.17, n = 27). Secular equilibrium was generally reached near the bottom depths in all basins except at stations 13 and 60 where the water samples were either enriched in $^{210}Po_t$ ($^{210}Po_t/^{210}Pb_t$ AR = 1.58 ± 0.16) or depleted in $^{210}Po_t$ ($^{210}Po_t/^{210}Pb_t$ AR = 0.50 ± 0.12), respectively.

~~Total ^{210}Po activities ($^{210}Po_t$) in all samples ranged from 2.2 to 16.4 dpm 100 L^{-1} and the mean $^{210}Po_t$ for all samples was 8.8 ± 2.4 dpm 100 L^{-1} (n = 198, Fig. 2). The corresponding total ^{210}Pb activities ($^{210}Pb_t$) were between 2.1 and 20.6 dpm 100L^{-1} with a mean value of 10.0 ± 3.0 dpm 100 L^{-1} (n = 198).~~

~~The mean $^{210}Po_t/^{210}Pb_t$ activity ratio (AR) of all samples was 0.92 ± 0.28 (Fig. 2, n = 198). When considering different basins separately, there is a tendency of decreasing $^{210}Po_t/^{210}Pb_t$ AR~~

~~from the Western European Basin (1.10 ± 0.35) westwards to the Iceland Basin (0.90 ± 0.19) and the Irminger Sea and the Labrador Sea (0.80 ± 0.18 and 0.83 ± 0.21, respectively). For all regions, within the mixed layer and euphotic zone (15 – 47 m), significant deficits of $^{210}Po_t$ (0.80 ± 0.20, n = 40) were observed (Fig. 3). $^{210}Po_t$ had enrichments below the surface at some depths at stations 1, 13, and 21 (Fig. 2) where the sub-surface $^{210}Po_t$ excesses were much larger than the surface depletion. In the depth below the surface to ~ 1500 m in the Iceland Basin, the Irminger Sea, and the Labrador Sea, the water samples still indicated a ^{210}Po deficiency (0.84 ± 0.17, n = 27). Secular equilibrium was generally reached near the bottom depths in all basins except at stations 13 and 60 where the water samples were enriched ($^{210}Po_t/^{210}Pb_t$ AR = 1.58 ± 0.16) and depleted ($^{210}Po_t/^{210}Pb_t$ AR = 0.50 ± 0.12) in $^{210}Po_t$, respectively. Secular equilibrium was also observed at some shallow depths (i.e. 80 m at station 44) and even in surface waters (i.e. 15 m at station 38).~~

3.2 Particulate ^{210}Po and ^{210}Pb activities

Small particulate ^{210}Po ($^{210}Po_s$) activities varied in a wide range from 0.08 to 4.82 dpm 100L^{-1} (mean: 0.76 ± 0.63 dpm 100L^{-1}, n = 81), about 83% of the values in the small particles were lower than 1.0 dpm 100L^{-1} with higher $^{210}Po_s$ values generally observed in the surface samples (Fig. 4, Table S2). The range of small particulate ^{210}Pb ($^{210}Pb_s$) activities was 0.07 to 2.89 dpm 100L^{-1} (mean: 0.56 ± 0.46 dpm 100L^{-1}, n = 81). 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 (Fig. 4). This has been seen in the North Atlantic along the GEOTRACES GA03 transect (Rigaud et al., 2015). The mean $^{210}Po_s/^{210}Pb_s$ activity ratio (AR) was 1.43 ± 0.96 in the surface waters (n = 14, ≤ 47 m), and 1.57 ± 0.90 with all samples included (n = 81, 8 – 3440 m). While most surface observations had an AR of $^{210}Po_s/^{210}Pb_s$ higher than unity, 5 surface samples at stations 69 and 77 showed an enrichment of ^{210}Pb activity over ^{210}Po ($^{210}Po_s/^{210}Pb_s$ AR: 0.62 ± 0.18).

Large particulate ^{210}Po ($^{210}Po_l$) activities ranged from 0.01 to 0.83 dpm 100L^{-1} with a mean of 0.10 ± 0.12 dpm 100L^{-1} (n = 59, Fig. 5, Table S2). The range of ^{210}Pb activity in the large particles ($^{210}Pb_l$) was from 0.02 to 0.67 dpm 100L^{-1} (mean: 0.12 ± 0.14 dpm 100L^{-1}, n = 59). The highest $^{210}Po_l$ and $^{210}Pb_l$ values were found at 30 m at station 26. The mean $^{210}Po_l/^{210}Pb_l$ activity ratio (AR) was 1.09 ± 1.54 in the surface waters (n = 14, ≤ 47 m), and 1.06 ± 0.86 when all data were considered (n = 59, 8-800 m). There were 17% of the samples with a depletion of ^{210}Po activity

relative to ^{210}Pb activity in large particles (mean AR: 0.49 ± 0.23), particularly in surface waters from the western section. We address this issue further in sections 4.2 and 4.3.

The percentages of total ^{210}Po activity in the small and large particles ranged from 0.9 to 46.7% (mean: 8.0 ± 6.7%) and from 0.1 to 8.9% (mean: 1.2 ± 1.5%), respectively. The percentage of total ^{210}Pb activity ranged from 0.7 to 21.4% (mean: 4.9 ± 3.8%) and from 0.2 to 5.9% (mean: 1.1 ± 1.2%) in the small and large particulate phase, respectively. These values revealed that both radionuclides were predominantly present in the dissolved phase along this transect, as is commonly found in the ocean. The particulate percentages reported here are similar to the values reported from the F.S. "Meteor" cruise 32 in the North Atlantic (Bacon et al., 1976) and along the North Atlantic GA03 transect (Rigaud et al., 2015).

We then combined radionuclide activity on the small and large particles from the same depth as the total particulate activity. There were 56 samples in total (surface to 800 m) and 41 of them were from the upper 200 m. Most of the total particulate ^{210}Po ($^{210}Po_p$) and ^{210}Pb ($^{210}Pb_p$) activity was on the small particles, with 86% of $^{210}Po_p$ and 80% of $^{210}Pb_p$ on the small size fraction (data not shown). The total particulate ^{210}Po and ^{210}Pb AR ($^{210}Po_p/^{210}Pb_p$) had the same mean as that of the small particulate ^{210}Po and ^{210}Pb AR ($^{210}Po_s/^{210}Pb_s$) (Welch Two Sample t-test, n = 56, $p = 0.1$), indicating that the values of the $^{210}Po_p/^{210}Pb_p$ activity ratios were driven by the small particles. While the majority of particulate matter was enriched in ^{210}Po ($^{210}Po_p/^{210}Pb_p$ AR> 1), there were 13 out of 56 total samples from various depths that were depleted in ^{210}Po relative to ^{210}Pb ~~While the majority of particulate matter was enriched in ^{210}Po ($^{210}Po_p/^{210}Pb_p$ AR> 1), there were some surface samples that were depleted in ^{210}Po relative to ^{210}Pb~~. The $^{210}Po_p/^{210}Pb_p$ activity ratios from this study are compared to the results from previous studies in various oceanic regimes in section 4.2.

4 Discussion

4.1 Total ^{210}Po and ^{210}Pb activities

The overall profiles of $^{210}Po_t$ and $^{210}Pb_t$ activities were different among basins (Fig. 2). The deficiencies of $^{210}Po_t$ activities with respect to $^{210}Pb_t$ activities in the surface samples from the Iceland Basin, the Irminger Sea, and the Labrador Sea were generally greater than those from the West~~ern~~ European Basin. Such disequilibria generally extended to the deep waters (1700 – 2950 m). In contrast, $^{210}Po_t$ activities in the West~~ern~~ European Basin were generally enriched relative to

$^{210}Pb_t$ activities from below the surface to the bottom of the profile. In the West~~ern~~ European Basin, the sub-surface $^{210}Po_t$ activity excess was much larger than the surface depletion, suggesting that some external source would be needed to maintain this excess ^{210}Po activity within the water column. One possible source of these sub-surface ^{210}Po activity excesses below 2000 m at stations 1 and 13 could be the North-East Atlantic Deep Water, lower ($NEADW_L$) which was the dominant water mass in the Iberian Basin from 2000 m to the bottom, and had a concentration of silicate up to 48 µmol kg^{-1} (García-Ibáñez et al., 2015). High activity of ^{210}Po in deep samples could be due to the dissolution of diatoms or herbivore feces (Cooper, 1952). As these particles sink and dissolve, ^{210}Po activity may have been preferentially released to the dissolved phase compared to ^{210}Pb activity (Bacon et al., 1976), leading to ^{210}Po excess observed in the deep waters at stations 1 and 13. For the sub-surface ^{210}Po activity excesses at station 1 between 400 and 1000 m where lateral inputs of particulate Fe from the margin was observed (Gourain et al., 2018), the likely process is diffusion of ^{210}Po from those particles originated from the margin and such excess could be transported westwards to station 13 by lateral advection. An alternative source of ^{210}Po activity excess between 50 and 250 m at stations 1 and 13 (Fig. 3) could be the eastern boundary upwelling along the coast of the Iberian Peninsula (García-Ibáñez et al., 2015). Even though no strong upwelling events were revealed from temperature and density profiles during the cruise, northerly winds favoring upwelling were recorded 2 – 3 months before the sampling (Shelley et al., 2017). The deep water may have excess ^{210}Po activity due to the remineralization of sinking particles. The upwelling of this water mass prior to the sampling date could maintain such sub-surface excess ^{210}Po activity. Similar findings have been reported in the Cariaco Trench for the upper 300 m of the water column by Bacon et al. (1980a).~~One possible source of these sub-surface ^{210}Po activity excesses could be the eastern boundary upwelling along the coast of the Iberian Peninsula (García-Ibáñez et al., 2015). Even though no strong upwelling events were revealed from temperature and density profiles during the cruise, northerly winds favoring upwelling were recorded 2 – 3 months before the sampling (Shelley et al., 2017). The deep water may have excess ^{210}Po activity due to the remineralization of sinking particles. The upwelling of this water mass prior to the sampling date could maintain excess ^{210}Po activity in the water column if the previous export of ^{210}Po activity was large enough. Similar findings have been reported in the Cariaco Trench by Bacon et al. (1980a).~~

As atmospheric deposition is the main source of ^{210}Pb to the water column (e.g. Masqué et al., 2002), we divided the GA01 transect into a western section (stn. 44 – 77) and an eastern section (stn. 1 – 38) based on atmospheric deposition boxes described in Shelley et al., (2017). Total atmospheric deposition fluxes of a suite of aerosol-sourced trace metals (TEs) were ~~all~~ reported to be higher in the east than the west for 18 out of 19 TEs (Shelley et al., 2017). However, a two sample t-test revealed a greater mean of $^{210}Pb_t$ activity in surface waters in the western than in the eastern section ($p < 0.02$, mean: 12.1 vs. 10.4 dpm 100 L^{-1}), despite the fact that ^{210}Pb is usually associated with aerosols. Even though the direct input of atmospheric ^{210}Pb may be larger in the east (assuming it behaves like the other trace metals, but without aerosol ^{210}Pb data we cannot confirm this), alternative inputs of ^{210}Pb from freshwater (e.g., sea ice processes and meteoric water) could be a greater source of ^{210}Pb activity to the west. The freshwater sources over the Greenland shelf and slope have been identified by Benetti et al. (2017), and were believed to be an important source of Fe (Tonnard et al., in review) and Al (Menzel-Barraqueta et al., in review) off of Greenland during this cruise. This ~~unexpected~~ result highlights the need in the future to measure ^{210}Pb activity simultaneously in the atmospheric and local freshwater sources in order to account for all source terms.

4.2 Total particulate ^{210}Po/^{210}Pb AR

A proposed explanation for the depletion of ^{210}Po activity relative to ^{210}Pb activity (AR <1) in some particles is effective recycling, commonly characterized by a subsurface excess of dissolved ^{210}Po activity released from enriched particles leaving the surface. Bacon et al. (1976) suggested that the efficiency of this recycling could reach up to 50%, while there is no significant concurrent release of ^{210}Pb activity in the water column. Laboratory studies have found the release rate of ^{210}Po in marine particulate matter to be significant; for example, 41% of the ^{210}Po activity in euphausiid fecal pellets was released over 5 days as presented in Heyraud et al. (1976). An alternative explanation for the depletion of ^{210}Po activity in particles is their lithogenic origin. ^{210}Po/^{210}Pb AR in lithogenic particles was reported to be similar to or less than unity (Nozaki et al., 1998; Tateda et al., 2003). In addition, the AR < 1 observed at station 1 (120, 250, and 550 m) could be associated with lithogenic particles from the Iberian Margin where 100% of the particulate Fe (PFe) had a lithogenic origin while the lithogenic contribution to PFe at other stations was smaller (Gourain et al., 2018)~~The AR < 1 observed at station 1 (120, 250, and 550 m)~~

~~could be associated with lithogenic particles from the Iberian Margin where the lithogenic contribution to particulate and dissolved Fe and dissolved Al were reported to be significant (Gourain et al., 2018; Menzel Barraqueta et al., in review)~~.

The time-series chlorophyll-a concentrations (8-day composite, https://oceancolor.gsfc.nasa.gov) from January to July 2014 at each station revealed bloom conditions about 4 months prior to the sampling time (~~Fig. 4~~ Fig. 6). We estimated the days since the last bloom began prior to the sampling date for each station (Table 1) and put these data into the context of the low $^{210}Po_p/^{210}Pb_p$ AR (< 1) in the total particles > 1 μm (Fig. 7). Eight stations had total particulate samples with $^{210}Po_p/^{210}Pb_p$ AR lower than unity from either shallow or deep waters. Specifically, when the time since the last bloom began was relatively short (24 – 47 d) the samples with $^{210}Po_p/^{210}Pb_p$ AR < 1 were observed in the shallow waters (10 – 60 m). In contrast, as longer time (50 – 74 d) passed since the last bloom, the depths at which samples had $^{210}Po_p/^{210}Pb_p$ AR < 1 were found to be much deeper (120 – 500 m). The results indicated that post-bloom particles could be recycled for weeks in shallow depths and take weeks to months to sink to deeper waters.

The averages of $^{210}Po_p/^{210}Pb_p$ AR within the upper 200 m water column were put into a global context with previously reported results (~~Fig. 5~~ Fig. 8). Total particulate $^{210}Po/^{210}Pb$ AR in the open ocean in previous studies (e.g., Equatorial/western Pacific, Bellingshausen Sea, BATS, Labrador Sea) were generally greater than unity. In contrast to the open ocean, the data show a distinct trend of depletion of relative ^{210}Po activity in marine particles from the shallow seas of the high latitude northern hemisphere. The lowest total particulate $^{210}Po/^{210}Pb$ AR values (Table 2, 0.4 – 0.5) were found in the ~~central Arctic and~~ Chukchi shelf (He et al., 2015) and other seas from the Eurasian sector (Barents, Kara and Laptev Seas) but also in central Arctic (Friedrich, 2011). Previous studies have observed depletion of relative ^{210}Po activity in nearshore particles in the Yellow Sea (Hong et al., 1999), in the turbid waters off of western Taiwan (Wei et al., 2012), on the shelf of Woods Hole, MA (Rigaud et al., 2015), and now in the margin station off St. John's, Canada (this study). The previous authors attributed the relative depletion of particulate ^{210}Po activity in the nearshore waters to the terrestrial origin/riverine input of particles with a low $^{210}Po/^{210}Pb$ AR. This may partially explain low activity ratios in the samples from the shelf of the Arctic Ocean as well, since it receives ~ 10% of global river runoff and is the most riverine-influenced of all of the world's oceans (Opsahl et al., 1999; Carmack et al., 2006). The Arctic

Basin, similarly, had ~~wide spread~~ widespread deficits of particulate ^{210}Po activity in the upper water column during the sea-ice minimum in 2007 (Roca-Marti et al., in review). Besides shelf particles, ~~The~~ the authors suggest~~ed~~ that other particle types could also play a role in lowering the particulate AR, including sea-ice sediments, remineralized material, fecal pellets, and picoplankton aggregates.

4.3 Relationship between total particulate ^{210}Po/^{210}Pb AR and AOU

AOU is a time-integrated measure of the amount of oxygen removed during the biogeochemical processes (e.g. respiration, remineralization, oxidation) in the ocean interior. Therefore, AOU is a product of apparent oxygen utilization rate (AOUR) and the age of water mass (e.g. Stanley et al., 2012), i.e. high AOU could be due to either intense biogeochemical processes that have occurred in a short period of time (young water mass) or weaker processes over a longer period of time (old water mass). Consequently, the rate of these biogeochemical processes and time (water mass age) would have different/similar impacts on the $^{210}Po_p/^{210}Pb_p$ AR value depending on the initial AR in the particles and the natural of the particles. ~~Apparent oxygen utilization (AOU = $O_{2\ saturation}$ – $O_{2\ measured}$), the amount of oxygen that has been consumed by remineralization of exported organic matter in the water column, can be used to indicate the intensity of particle recycling (Ito et al., 2004; Duteil et al., 2013). While AOU is generated both by water mass ageing and concomitant biological oxygen consumption (e.g. Ito et al., 2004; Sonnerup et al., 2015), the two components of AOU would be predicted to have opposite impacts on the $^{210}Po_p/^{210}Pb_p$ AR value.~~ For example, the $^{210}Po_p/^{210}Pb_p$ AR would tend to increase with time if the initial AR is < 1 because particulate ^{210}Po activity would increase from the decay of ^{210}Pb and trend towards secular equilibrium ($^{210}Po_p/^{210}Pb_p$ AR = 1), and to decrease with time if the initial AR is > 1 as the original excess of particulate ^{210}Po activity would disappear after 7 half-lives of ^{210}Po.~~old particles would tend to have a higher $^{210}Po_p/^{210}Pb_p$ activity ratio (closer to 1) because particulate ^{210}Po activity would increase from the decay of ^{210}Pb within mineral lattices and trend towards secular equilibrium ($^{210}Po_p/^{210}Pb_p$ AR = 1)~~. In contrast, oxygen consumption due to bacterial remineralization would preferentially release ^{210}Po activity from particles into the dissolved pool (e.g. Stewart et al., 2008), leading to a lower $^{210}Po_p/^{210}Pb_p$ AR in those particles.

The combination of average $^{210}Po_p/^{210}Pb_p$ AR and their corresponding average AOU in the upper 200 m at 40 stations from 4 independent studies, including ARK-XXII/2 (77.38 – 87.83 ºN,

n = 15) in the Arctic, BOFS (48.89 – 49.87 ºN, n = 7), GA03 (22.38 – 39.70 ºN, n = 7), and GA01 (this study, 40.33 – 59.80 ºN, n = 11) in the North Atlantic (see map in ~~Fig. 5~~ Fig. 8) suggests two distinct linear trends (~~Fig. 6~~ Fig. 9). When AOU was lower than 25 µmol kg^{-1}, the $^{210}Po_p/^{210}Pb_p$ AR was found to be greater than unity, together with a linear negative relationship (n = 27, R^2 = 0.5, $p < 0.001$) towards the AOU at 25 µmol kg^{-1}. In contrast, AOU values greater than 25 µmol kg^{-1} were coincident with a $^{210}Po_p/^{210}Pb_p$ AR < 1, and there was a linear positive relationship (n = 12, R^2 = 0.4, p = 0.03) towards the highest AOU values measured. The two contradictory linear trends likely reflect the nature of the particles. For example, the observation of $^{210}Po_p/^{210}Pb_p$ AR > 1 with AOU < 25 µmol kg^{-1} may suggest relatively fresh/organic particles in the young water mass. When AOU increases either due to water mass aging or higher AOUR, the $^{210}Po_p/^{210}Pb_p$ AR decreases with a slope of -0.17 ± 0.04. On the other hand, refractory/lithogenic particles may be suggested by the observation of $^{210}Po_p/^{210}Pb_p$ AR < 1 with AOU > 25 µmol kg^{-1}. For those particles, increasing in AOU either due to water mass aging or higher AOUR would change the $^{210}Po_p/^{210}Pb_p$ AR to a much lesser degree than that for organic particles with a slope of 0.008 ± 0.003. ~~The two contradictory linear trends likely reflect the opposite impacts of the two components (water mass aging and remineralization) of AOU on $^{210}Po_p/^{210}Pb_p$ AR. This suggests that the variation in the $^{210}Po_p/^{210}Pb_p$ AR was mainly driven by remineralization processes under the condition of AOU < 25 µmol kg^{-1}, lowering the total particulate activity ratio; whereas the decay of ^{210}Pb into ^{210}Po towards secular equilibrium may dominate when AOU was > 25 µmol kg^{-1}, leading to an increase in $^{210}Po_p/^{210}Pb_p$ AR.~~ This explanation, however, appears to only hold for the high latitude Northern Hemisphere where $^{210}Po_p/^{210}Pb_p$ activity ratios were generally lower than those in the other oceanic settings (~~Fig. 5~~ Fig. 8). In the high latitude Southern Hemisphere near Antarctic (e.g., ANT-X/6), for example, there is no apparent relationship between $^{210}Po_p/^{210}Pb_p$ activity ratios and AOU. This relationship (or lack thereof) deserves more study in the future.

4.4 ~~Small particles, sorption, and calculating POC export~~ Relationship among small particles, adsorption, and scavenging

The partitioning coefficient, K_d (L kg^{-1}), has been used to describe the particle adsorption behavior of radionuclides. It is defined as the ratio of the adsorbed radionuclide activity (A_p, dpm $100L^{-1}$) to the dissolved radionuclide activity (A_d, dpm $100L^{-1}$), normalized by the suspended particulate matter concentration (SPM, µg L^{-1}):

$$K_d = \frac{A_p}{A_d} \times \frac{1}{SPM} 10^9 \qquad (1)$$

Owing to the different biological and chemical behaviors of ^{210}Po and ^{210}Pb, the interpretation of measured K_d for ^{210}Po (K_d(Po)) may not be as clear as that for ^{210}Pb (K_d(Pb)). 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.

In this study, the size-fractionated data of both radionuclide activity and SPM allowed us to calculate the partitioning coefficients for both radionuclides on small and total particles. The dissolved radionuclide activity was calculated as the difference between total and particulate activity. The coefficients for the small particulate and the total particulate phases were normalized by the SPM in the small and total particulate phases, respectively. We present only the coefficients for the small particulate phases ($K_d(Po)_s$, $K_d(Pb)_s$) and the total particulate phases ($K_d(Po)_p$, $K_d(Pb)_p$) because most of the particulate activity (> 80%) was associated with the small particles along the GEOVIDE transect, and most conceptualized scavenging models consider either the two-box model (dissolved – total particulate phases, i.e. $K_d(Po)_p$) or the three-box model (dissolved – small – large, i.e. $K_d(Po)_s$) (Clegg and Whitfield, 1990; 1991; Rigaud et al., 2015) and thus activity is concentrated from the dissolved phase to the total or small particles.

The average values of K_d(Po) was 1.6 times of those of K_d(Pb) in both small and total particulate phases, suggesting a higher affinity with particles for ^{210}Po with respect to ^{210}Pb, which is commonly observed in the global ocean (Bacon et al., 1988; Hong et al., 1999; Masqué et al., 2002; Wei et al., 2014; Tang et al., 2017). The K_d values for the small particulate phase were slightly higher than those for the total particulate phase but overall these values were very similar for both radionuclides (Fig. 10), suggesting that adsorption/scavenging of radionuclides was driven by small particles along the transect. In addition, there are increasing studies which argue that small particles can form aggregates that sink, and their contribution to carbon export could be larger than previously thought (e.g. Richardson and Jackson, 2007; Lomas and Moran, 2011; Amacher et al., 2013; Puigcorbé et al., 2015). We, therefore, recommend combining the activities of both small and large particles into a total particulate fraction in order to explain total ^{210}Po/^{210}Pb disequilibria in the surface waters, and utilizing the characteristics of the total particles (instead of just the large particles) in the estimation of the POC export fluxes (Tang et al., companion paper submitted to this volume).

Traditionally, large particles collected by in-situ filtration with pumps, most commonly defined as particles larger than 53 or 70 μm, were assumed to dominate the sinking flux (Dugdale and Goering, 1967; Bishop et al., 1977; Fowler and Knauer, 1986; Honjo et al., 1992; Walsh and Gardner, 1992) such that the composition (POC/^{210}Po) of the large particle size class was used to convert ^{210}Po fluxes into POC export (e.g. Friedrich and Rutgers van der Loeff, 2002; Cochran and Masqué, 2003; Murray et al., 2005; Stewart et al., 2010; Roca-Martí et al., 2016). Given that the true size spectrum of sinking particles for the timescale relevant to the ^{210}Po/^{210}Pb method is unknown and the POC flux estimates are sensitive to the particulate POC/^{210}Po ratio, both small and large particles should be sampled for POC/^{210}Po due to the variability in the POC/^{210}Po ratio in different size classes (Hayes et al., in review).

~~The assumption that the largest particles dominate export in the ocean (e.g. Bishop et al., 1977; Fowler and Knauer, 1986; Michaels and Silver, 1988; Honjo et al., 1992; Walsh and Gardner, 1992) has been challenged by increasing studies which argue that small particles can form aggregates that sink, and their contribution to carbon export could be larger than previously thought (e.g. Richardson and Jackson, 2007; Lomas and Moran, 2011; Amacher et al., 2013; Puigcorbé et al., 2015).~~

~~We investigated the role of small phytoplankton to carbon export along the GA01 transect via investigation of pigments and *in-situ* primary production. The fraction of pigment-based size classes suggested a significant contribution of small particles (nano-phytoplankton: 2 – 20 μm 60%, pico-phytoplankton: < 2 μm, 13%) to primary production in the eastern section while larger particles (micro-phytoplankton: > 20 μm, 60%) may have dominated production in the western section of the GA01 transect (Tonnard et al., in prep.). The rate of primary production in the eastern section (mean: 99 ± 50 mmol C m^{-2} d^{-1}), however, was similar to that in the west (mean: 93 ± 58 mmol C m^{-2} d^{-1}) (data not shown). While we do not have direct evidence of small particles sinking, we are making an assumption that our study sites behave as the above cited papers have seen elsewhere. Therefore, a possible link between small particles and production, and possibly export (proportional to their role in production according to Richardson and Jackson, 2007), may exist along the transect.~~

~~The partitioning coefficient, K_d (L kg^{-1}), has been used to describe the particle adsorption behavior of radionuclides. It is defined as the ratio of the adsorbed radionuclide activity (, dpm $100L^{-1}$) to~~

the dissolved radionuclide activity (, dpm 100L^{-1}), normalized by the suspended particulate matter concentration (SPM, μg L^{-1}):

$$K_d = \frac{A_p}{A_d} \times \frac{1}{SPM} 10^9 \quad (1)$$

Owing to the different biological and chemical behaviors of ^{210}Po and ^{210}Pb, the interpretation of measured K_d for ^{210}Po (K_d(Po)) may not be as clear as that for ^{210}Pb (K_d(Pb)) (i.e. K_d(Po) also takes the fraction of absorbed ^{210}Po into account, Tang et al., 2017). As such, it would be more appropriate to think of both K_d(Po) and K_d(Pb) as the intensity parameter for the radionuclide association with particles.

In this study, the size-fractionated data of radionuclide activity and SPM allowed us to calculate the partitioning coefficients for both radionuclides on small and large particles. We present only the coefficients for the small particulate phases ($K_d(Po)_s$, $K_d(Pb)_s$) and the total particulate phases ($K_d(Po)_p$, $K_d(Pb)_p$) because most of the particulate activity (> 80%) was associated with the small particles along the GEOVIDE transect, and most conceptualized scavenging models consider either the two-box model (dissolved – total particulate phases, i.e. $K_d(Po)_p$) or the three-box model (dissolved – small – large, i.e. $K_d(Po)_s$) (Clegg and Whitfield, 1990; 1991; Rigaud et al., 2015) and thus activity is concentrated from the dissolved phase to the total or small particles. The K_d values for the small particulate phase were slightly higher than those for the total particulate phase but overall these values were very similar for both radionuclides (Fig. 7). Combining the fact that adsorption/scavenging was in fact driven by small particles with the contribution of small phytoplankton to production, the importance of small particles to radionuclide export is suggested. We recommend combining the activities of both small and large particles into a total particulate fraction in order to explain total ^{210}Po/^{210}Pb disequilibria in the surface waters, and utilizing the characteristics of the total particles (instead of just the large particles) in the estimation of the POC export fluxes (Tang et al., in prep.).

5 Conclusions

In this study, we reported the vertical distribution of total and size-fractionated particulate ^{210}Po and ^{210}Pb activities in the North Atlantic during the GEOVIDE GA01 cruise. More than 90% of the radionuclide activity was found in the dissolved phase, while a small proportion was associated with particles in this transect. Total ^{210}Po activity was generally depleted relative to total ^{210}Pb activity in the upper 100 m due to the preferential adsorption of ^{210}Po activity by particles. Such

deficiencies of ^{210}Po activities generally extended to the deep waters at most of the stations. In the West European Basin, the excess of ^{210}Po activities at stations 1 and 13 in the North East Atlantic Deep Water was attributed to the release of ^{210}Po during dissolution of sinking biogenic particles.

There appear to be geographic differences in particulate ^{210}Po/^{210}Pb activity ratios measured during GEOVIDE and previous studies, with particularly low values in the high-latitude North Atlantic and Arctic. While this observation deserves more attention, we support previous suggestions that this is due to the terrestrial origin/riverine input of particles with a low ^{210}Po/^{210}Pb AR into the river-dominated shallow seas of the Arctic. The age of the particles and water masses as well as the importance of biogeochemical processes (e.g. respiration, remineralization) may also explain some of these observations, as there was a significant relationship between the total particulate activity ratio and AOU when both were measured in the North Atlantic (> 20 ºN) and Arctic Oceans.

Over 80% of the particulate radionuclide activity was on small particles, indicating that the scavenging of both radionuclides was driven by small particles. Therefore, we suggest considering the activities of ^{210}Po and ^{210}Pb from both small and large particles in order to study the water column ^{210}Po/^{210}Pb disequilibria and quantify POC export along the GA01 transect. This has been addressed in a companion paper in this issue. We recommend that both small and large particles should be sampled for POC/^{210}Po estimates for the application of the ^{210}Po/^{210}Pb method in future studies of POC export.

~~In this study, we reported the vertical distribution of total and size-fractionated particulate ^{210}Po and ^{210}Pb activities in the North Atlantic during the GEOVIDE GA01 cruise. More than 90% of the radionuclide activity was found in the dissolved phase, while a small proportion was associated with particles in this transect. Total ^{210}Po activity was generally depleted relative to total ^{210}Pb activity in the upper 100 m due to the assumed preferential adsorption and uptake of ^{210}Po activity by particles.~~

~~Over 80% of the particulate radionuclide activity was on small particles, and it appeared that the adsorption/scavenging of both radionuclides was driven by small particles. Considering this and the contributions of small phytoplankton to primary production (and possibly export), we suggest combining the activities of both ^{210}Po and ^{210}Pb from both small and large particles into a total particulate fraction (> 1 µm) in order to explain the water column ^{210}Po/^{210}Pb disequilibria and calculate POC export.~~

There appear to be geographic differences in particulate $^{210}Po/^{210}Pb$ activity ratios measured during GEOVIDE and previous studies, with particularly low values in the high-latitude North Atlantic and Arctic. While this observation deserves more attention, we support previous suggestions that this is due to the terrestrial origin/riverine input of particles with a low $^{210}Po/^{210}Pb$ AR into the river-dominated shallow basins of the Arctic. Considering the age of the particles and water masses as well as the importance of remineralization may also explain some of these observations, as there was a significant relationship between the total particulate activity ratio and AOU when both were measured in the high latitude North Atlantic and Arctic Oceans.

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Table 1. Biological characteristics of the water column determined by chlorophyll-a concentration (8-day composite) from ~~Fig. 4~~ Fig. 6, including the date when the last bloom began, the difference in chlorophyll-a concentration between the sampling time and last bloom peak, and the days since the last bloom. Activity ratios of $^{210}Po_p/^{210}Pb_p < 1$ and their corresponding depths are also shown. *NA* indicates that all samples from the corresponding depth range had $^{210}Po_p/^{210}Pb_p$ equal to or greater than 1 (no sample with $^{210}Po_p/^{210}Pb_p < 1$).

Station	Sampling date	The date last bloom began	Last bloom peak-current state	Days since last bloom	$^{210}Po_p/^{210}Pb_p < 1$	
					0-100 m	> 100 m
1	5/19/14	3/6/14	Large	74	*NA*	Yes (120, 250, 500 m)
13	5/24/14	4/7/14	Small	47	Yes (60 m)	*NA*
21	5/31/14	4/7/14	Large	54	*NA*	Yes (120 m)
26	6/4/14	4/15/14	Large	50	*NA*	Yes (400 m)
32	6/7/14	5/9/14	Small	29	*NA*	*NA*
38	6/10/14	5/17/14	Small	24	Yes (60 m)	*NA*
44	6/13/14	5/9/14	Small	35	*NA*	*NA*
60	6/18/14	5/17/14	Large	32	*NA*	*NA*
64	6/19/14	5/17/14	Small	33	Yes (30 m)	*NA*
69	6/22/14	5/25/14	Small	28	Yes (20, 30 m)	*NA*
77	6/26/14	5/25/14	Small	32	Yes (10, 20, 50 m)	*NA*

997 Table 2. ~~Global~~ The compilation of total particulate $^{210}Po/^{210}Pb$ activity ratios ($^{210}Po_p/^{210}Pb_p$) averaged in the upper 200 m, including
998 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)

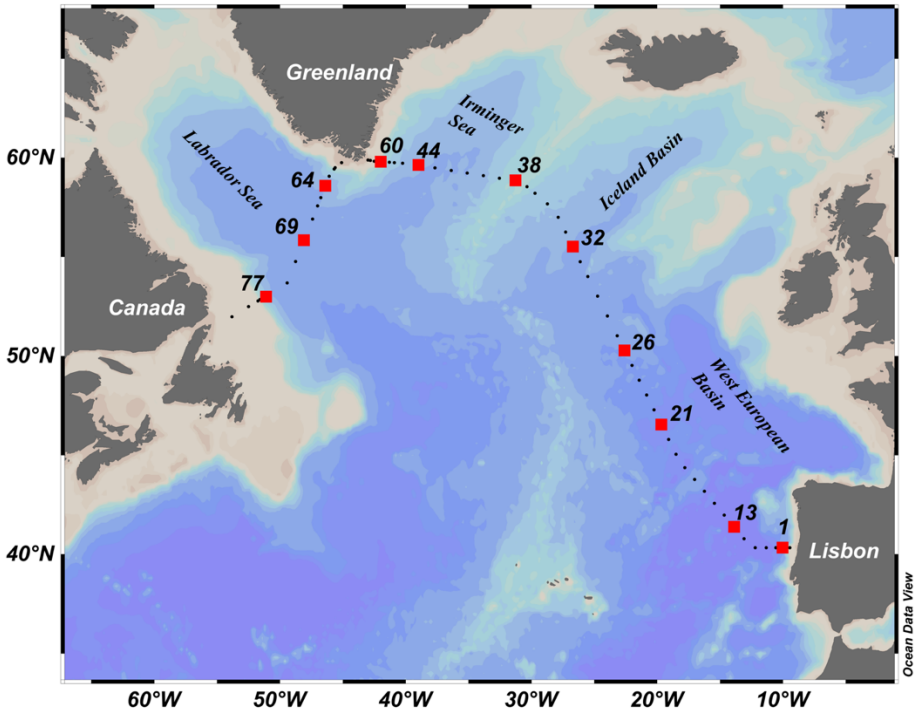

Fig. 1. Map of the GEOVIDE cruise track (black dots) and the 11 stations sampled for ^{210}Po and ^{210}Pb activity (red squares). Each sampling location is labeled with a station number. The sampling stations are divided into 4 regions (from east to west): West European Basin (stations 1, 13, 21, 26), Iceland Basin (stations 32, 38), Irminger Sea (stations 44, 60), and Labrador Sea (stations 64, 69, 77).

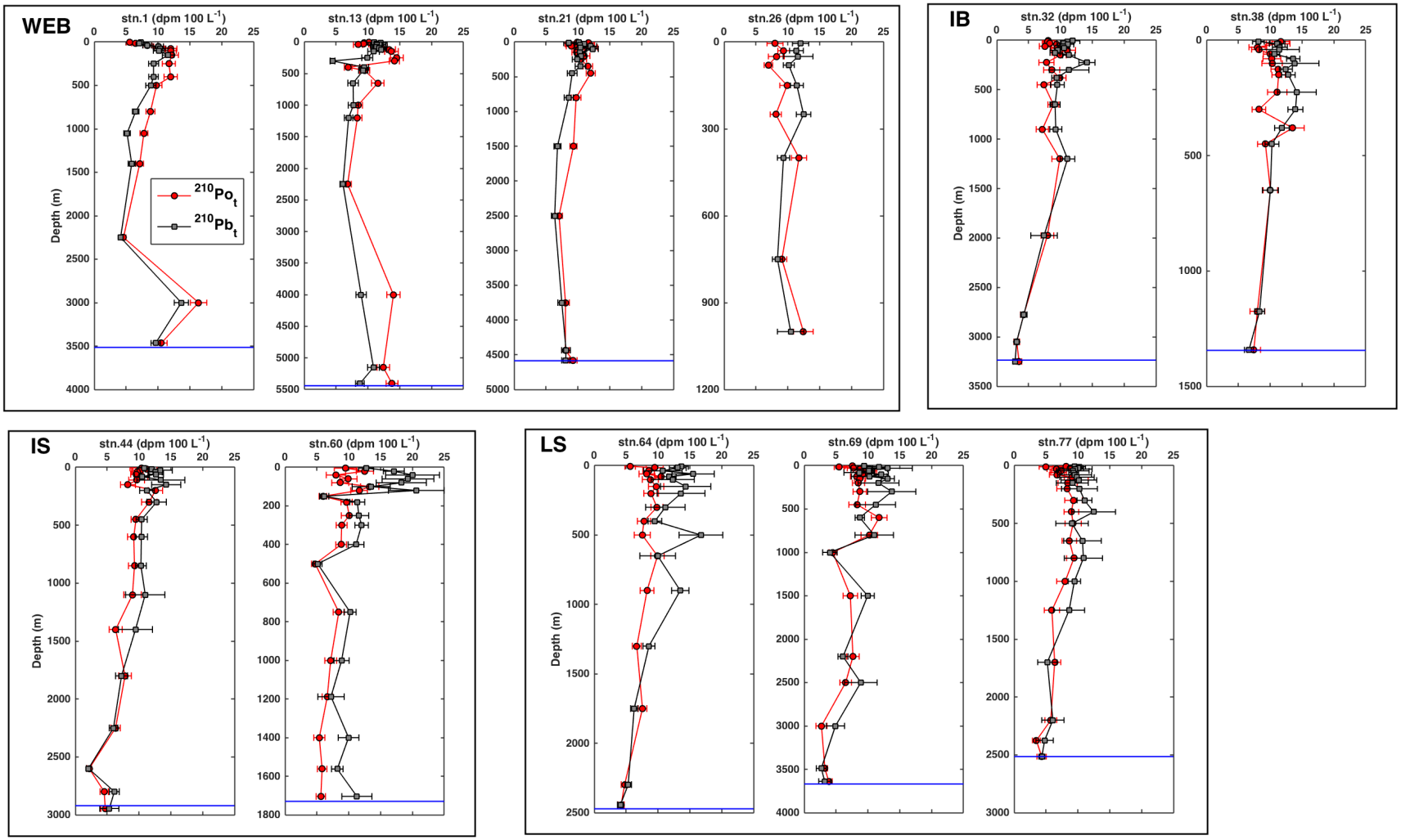

Fig. 2. The depth profiles of total ^{210}Po ($^{210}Po_t$, red circles) and ^{210}Pb activities ($^{210}Pb_t$, grey squares) along GEOVIDE section. The horizontal blue line is the bottom depth, which coincided with the deepest water sample except for station 26 which was sampled only down to 1000 m. Note that the depth scale for each plot may be different. The profiles are shown in the order of sampling date with

1012 the region indicated on the top left of each box: West~~ern~~ European Basin (WEB), Iceland Basin (IB), Irminger Sea (IS), Labrador Sea
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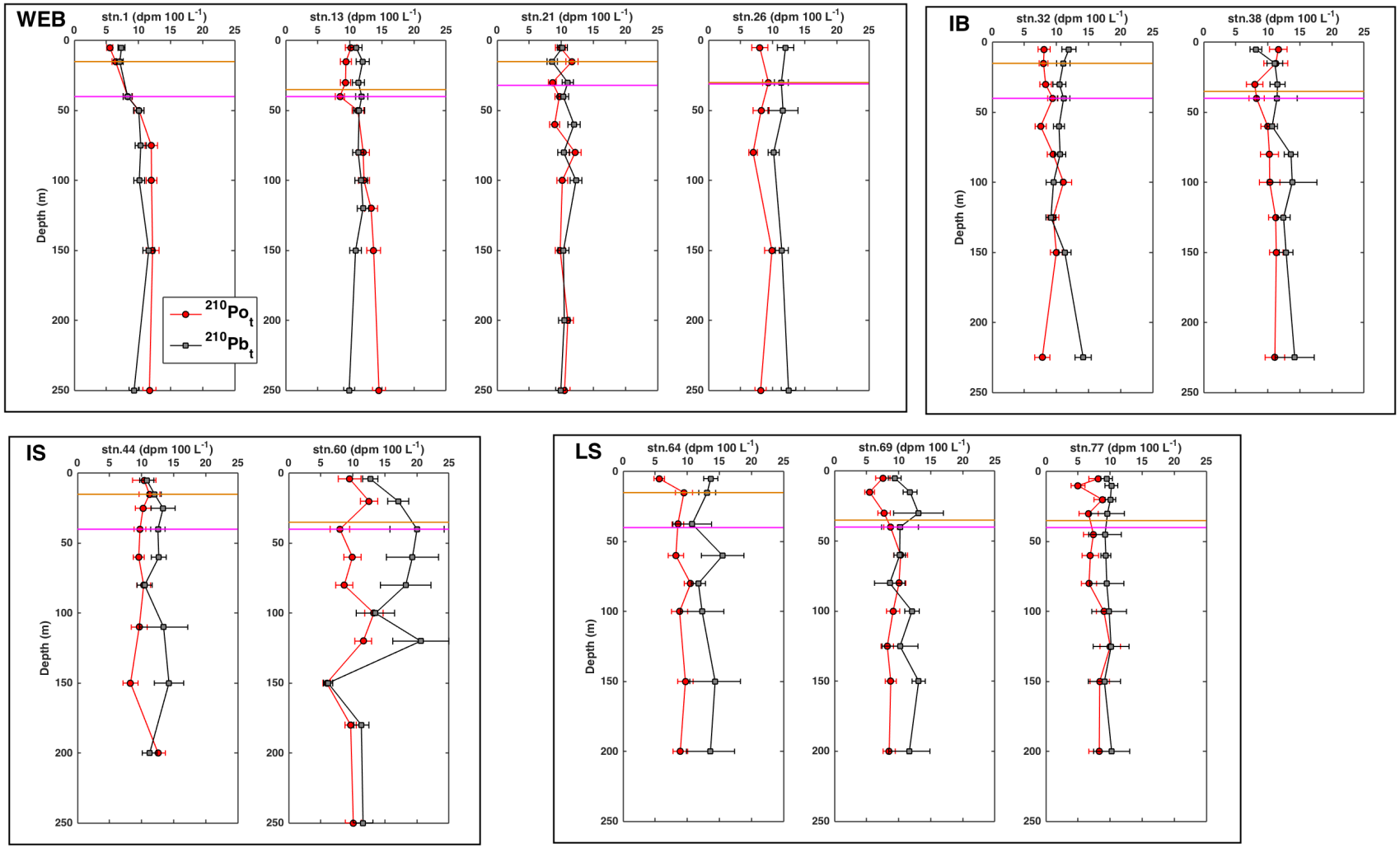

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1017 Fig. 3. ~~A closer look at only the zoom for the~~ The upper 250 m of the depth profiles of total ^{210}Po ($^{210}Po_t$, red circles) and ^{210}Pb

1018 activities ($^{210}Pb_t$, grey squares) along the GEOVIDE section. The horizontal orange and magenta lines denote the mixed layer depth

(MLD) and the base of the euphotic zone ($Z_{1\%}$), respectively. The depth profiles are shown in the order of sampling and grouped by region (refer to Fig. 2 for the text abbreviations).

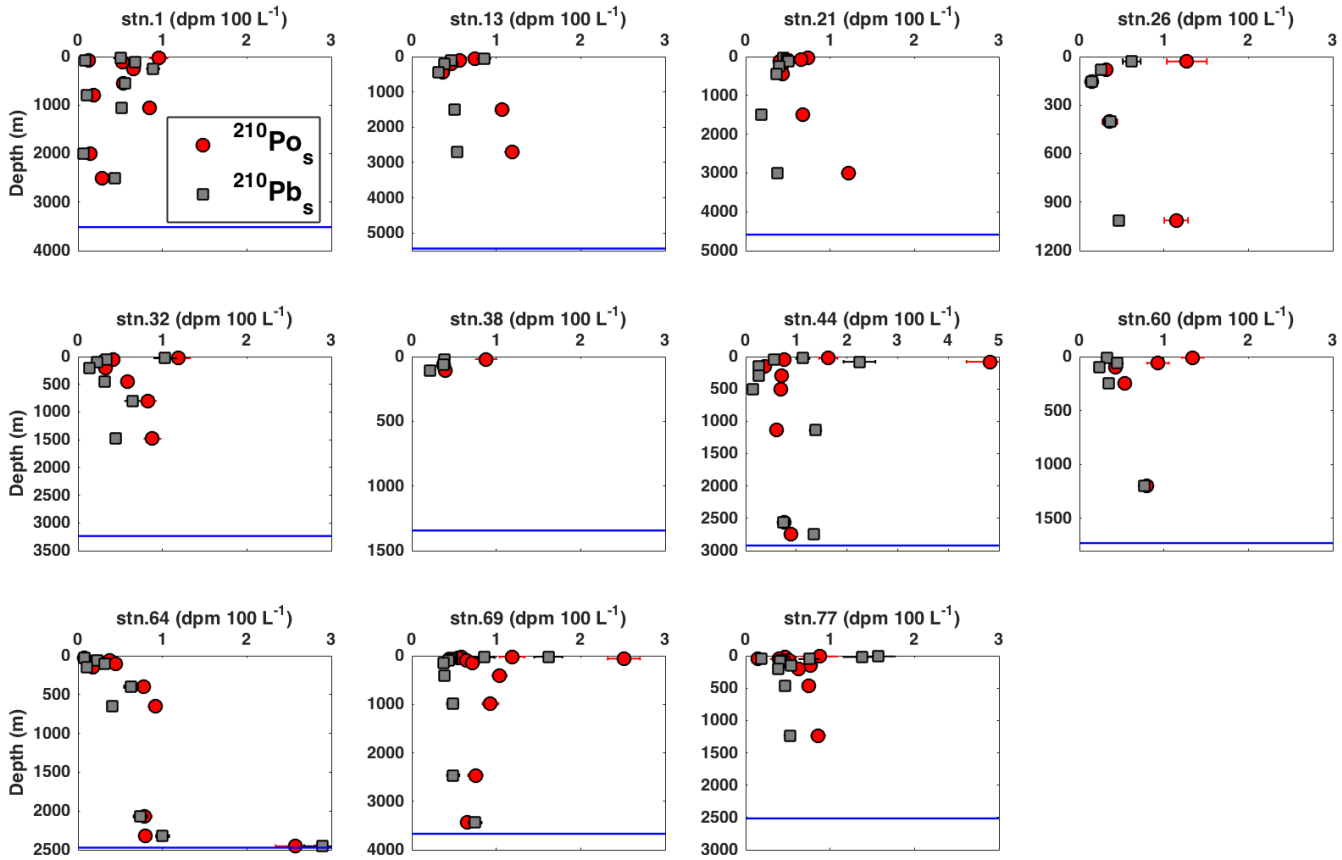


Fig. 4. Vertical profiles of the particulate ^{210}Po and ^{210}Pb activity in the small size fraction (1-53 µm, $^{210}Po_s$, $^{210}Pb_s$). Note that the depth scale may differ among plots, and the activity scale at Station 44 differs from the scale on all other plots. The horizontal blue line represents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

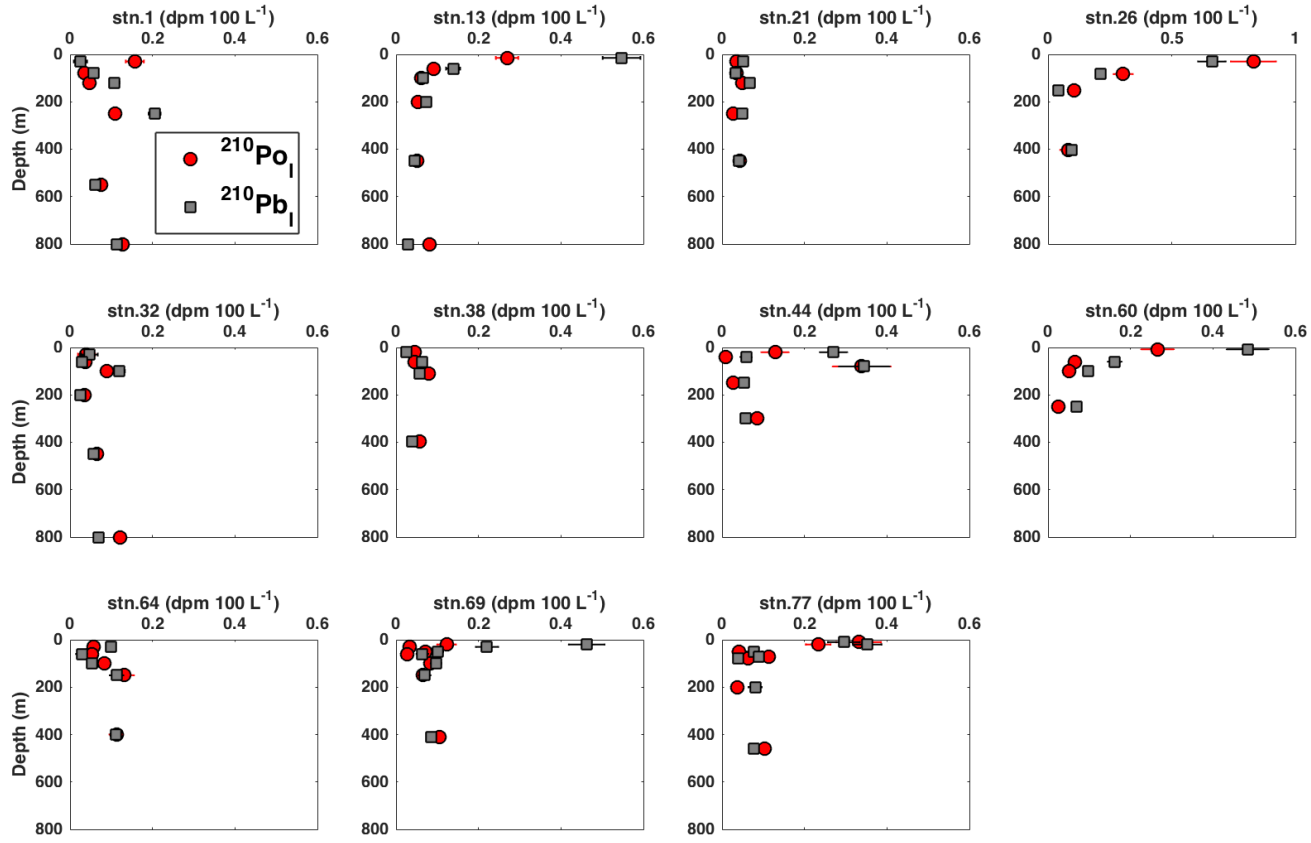
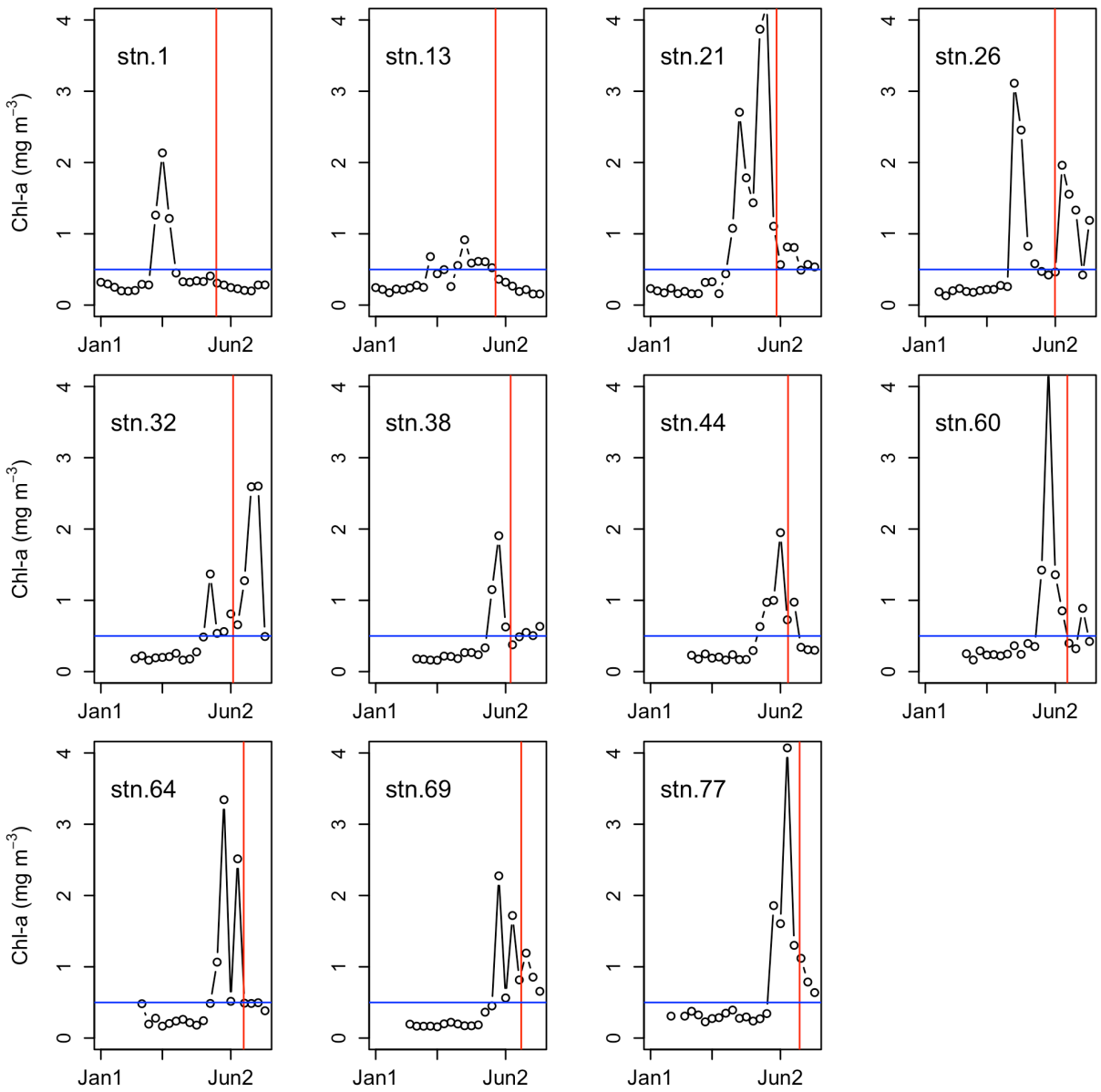
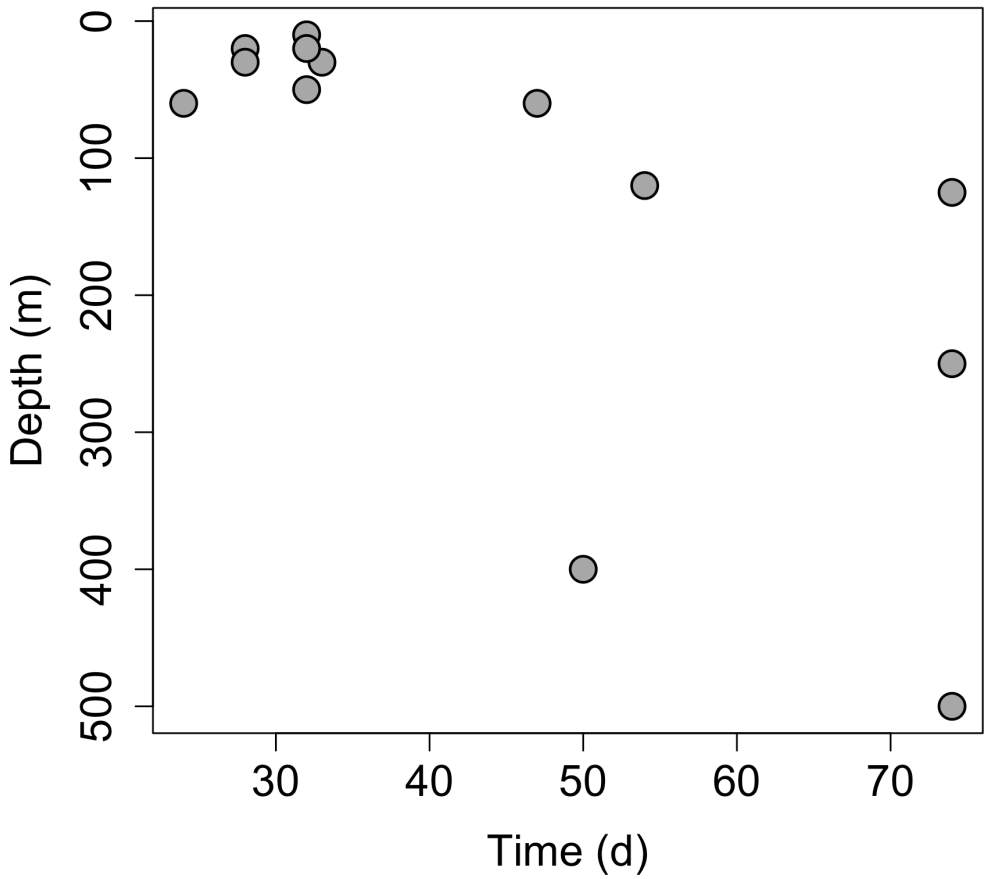


Fig. 5. The vertical profiles of the particulate ^{210}Po and ^{210}Pb activity in the large size fraction (> 53 μm, $^{210}Po_l$, $^{210}Pb_l$) in the top 800 m. Note that the activity scale at Station 26 differs from the scale on all other plots.



~~Fig. 4~~ Fig. 6. Time-series (January 1 – July 12, 2014) chlorophyll-a concentrations (8-day averages) from Aqua MODIS (https://oceancolor.gsfc.nasa.gov) at each station along the GA01 transect. The vertical red line denotes the sampling date at each station. The horizontal blue line denotes chlorophyll-a concentration of 0.5 mg m^{-3}. The time when chlorophyll-a concentration first exceeded 0.5 mg m^{-3} after the end of the last bloom defines the date when the next bloom began.



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1039 Fig.7. Depths at which the total particulate (> 1 μm) $^{210}Po/^{210}Pb$ activity ratio was lower than

1040 unity vs. the time since the last bloom (data is presented in Table 1).

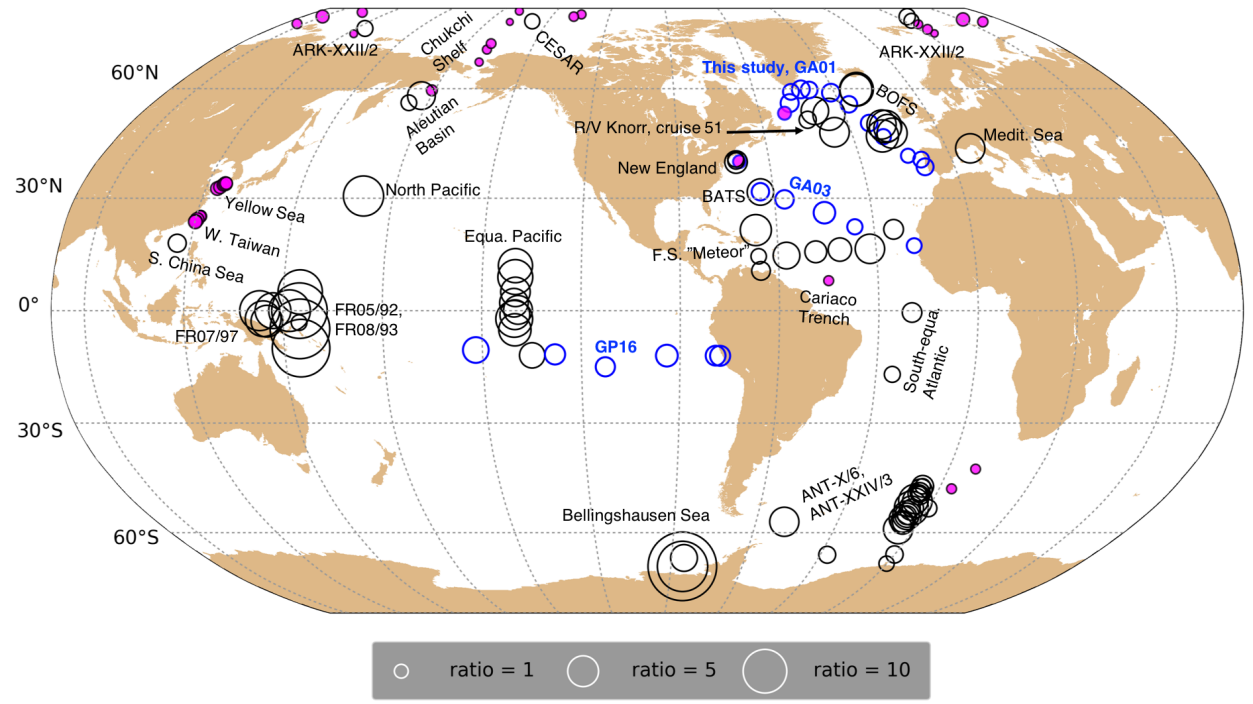
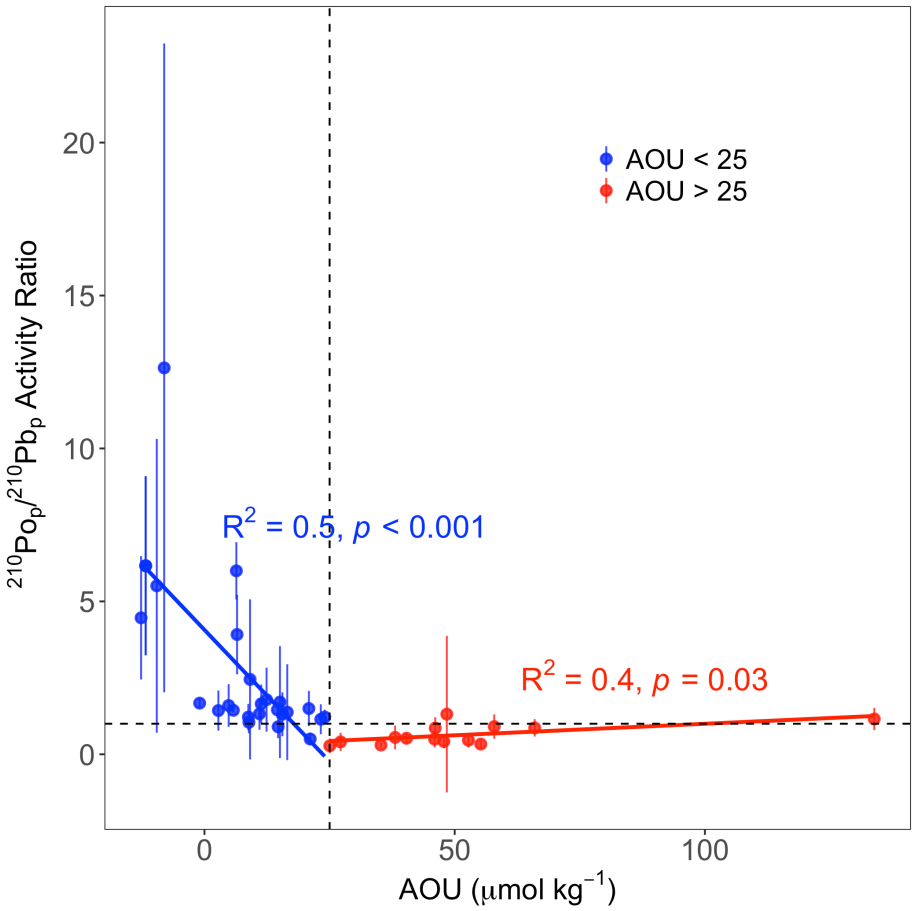
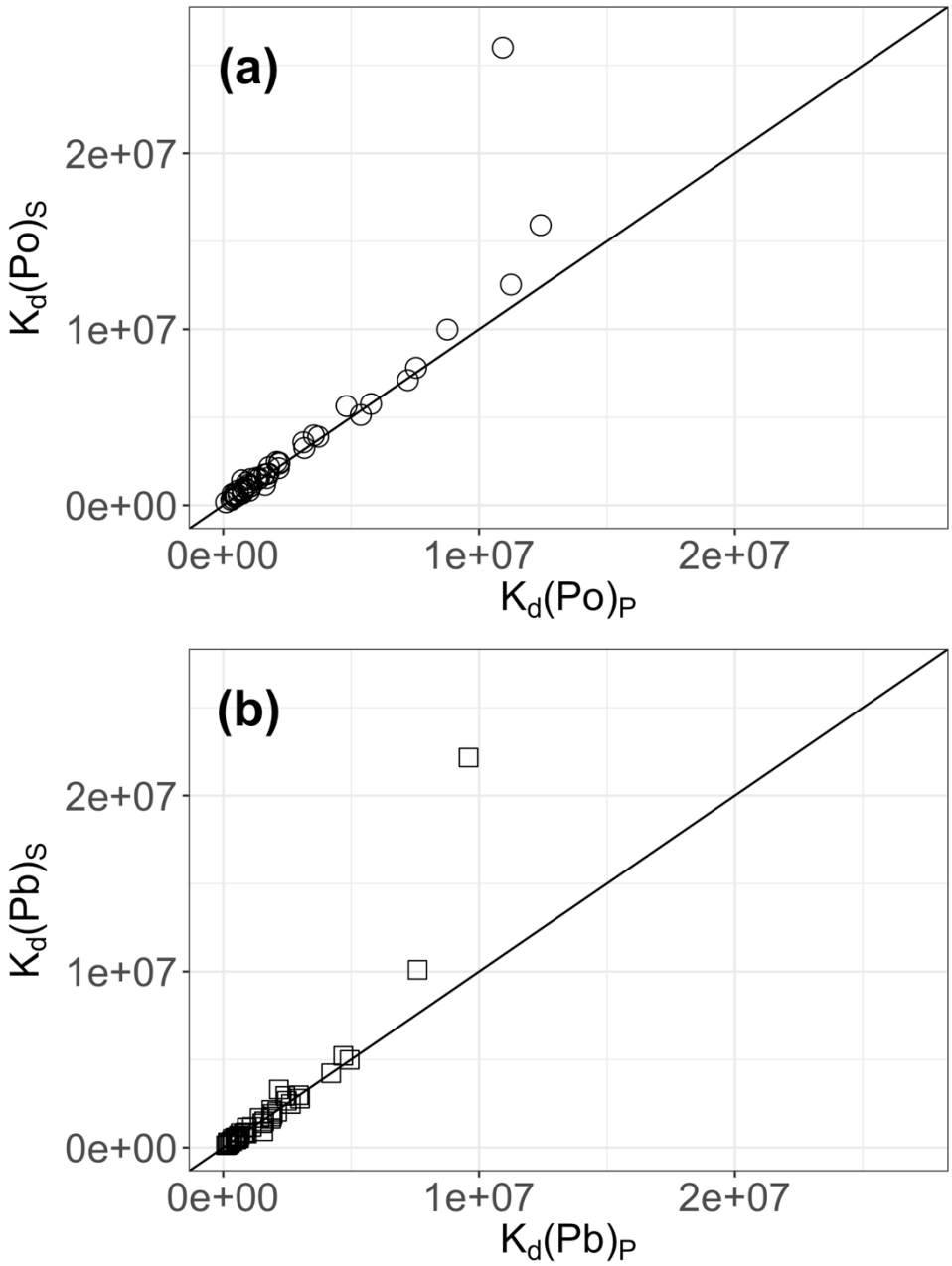


Fig. ~~5~~ Fig. 8. Comparison of particulate $^{210}Po/^{210}Pb$ activity ratios in the upper 200 m from this study and 20 previous studies (references in Table 2). Information about the study site, sampling date, method, and particle size of each study are shown in Table 2. The black circles represent data from previous studies while the ~~red~~ blue circles are the results from samples analyzed in the Stewart lab from three recent GEOTRACES transects (GA03, GP16, and this study, GA01 GEOVIDE). The filled ~~blue~~ magenta and open circles indicate activity ratios lower and higher than 1, respectively.



~~Fig. 6~~ Fig. 9. The relationship between AOU (µmol kg^{-1}) and total particulate $^{210}Po/^{210}Pb$ activity ratio ($^{210}Po_p/^{210}Pb_p$) from the upper 200 m in the northern hemisphere (> 22 ºN) investigated by a linear regression model (red and blue lines). The 40 stations include data from previous studies, ARK-XXII/2 (77.38-87.83 ºN, n = 15) in the Arctic, BOFS (48.89-49.87 ºN, n = 7), GA03 (22.38-39.70 ºN, n = 7), and this study, GA01 (40.33-59.80 ºN, n = 11) in the North Atlantic. The horizontal dashed line represents $^{210}Po_p/^{210}Pb_p$ AR = 1 and the vertical dashed line represents AOU = 25 µmol kg^{-1}. ~~Red~~ Blue circles denote ~~the average $^{210}Po_p/^{210}Pb_p$ > 1 and~~ AOU < 25 µmol kg^{-1}, while ~~blue~~ red circles ~~denote the average $^{210}Po_p/^{210}Pb_p$ < 1 and~~ AOU > 25 µmol kg^{-1}. ~~Data that are in neither category are denoted by the black circles.~~



~~Fig. 7~~ Fig. 10. Comparison of the partitioning coefficient (K_d) between the dissolved and small particulate phases (~~K_{ds}~~ $K_d(Po)_s$, $K_d(Pb)_s$) vs. between the dissolved and total particulate phases (~~K_{dp}~~ $K_d(Po)_p$, $K_d(Pb)_p$) for (a) ^{210}Po and (b) ^{210}Pb. The 1:1 line is indicated as the solid line in each plot.