

1     **Distributions of total and size-fractionated particulate  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities along the**  
2                     **North Atlantic GEOTRACES GA01 (GEOVIDE) transect**

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16 **Abstract**

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

33

34

## 35 1 Introduction

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

51 Polonium-210 ( $^{210}\text{Po}$ ,  $T_{1/2} = 138.4$  d) and its radioactive grandparent Lead-210 ( $^{210}\text{Pb}$ ,  $T_{1/2} =$   
52 22.3 y) are two non-conservative  $^{238}\text{U}$  decay series products. The GEOTRACES program has  
53 included both radionuclides in its TEIs list primarily due to  $^{210}\text{Po}$ 's enhanced bioaccumulation and  
54 the use of the  $^{210}\text{Po}/^{210}\text{Pb}$  pair as a proxy for assessing particle export in the upper ocean. The  
55 distribution of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  has been widely measured over the last several decades in the  
56 Atlantic (e.g. Bacon et al., 1976; Sarin et al., 1999; Rigaud et al., 2015; Ceballos-Romero et al.,  
57 2016), Pacific (e.g. Nozaki and Tsunogai, 1976; Murray et al., 2005; Verdeny et al., 2008), Indian  
58 (e.g. Cochran et al., 1983; Sarin et al., 1994; Subha Anand et al., 2017), Arctic (e.g. Moore and  
59 Smith, 1986; He et al., 2015; Roca-Martí et al., 2016) and Southern Oceans (e.g. Shimmield et al.,  
60 1995; Friedrich and Rutgers van der Loeff, 2002). However, since the data reported by Bacon et  
61 al., (1980b) at the Labrador Sea stations (47.8 – 53.7 °N), there are few studies of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$   
62 activity in the North Atlantic at latitudes greater than 40 °N. The GEOVIDE cruise, which targeted  
63 the North Atlantic from 40 °N to 60 °N, provided an opportunity to fill this data gap.

64 Besides ascertaining the distribution of the natural radionuclides under specific geographic  
65 conditions, this project aimed to answer questions about their biogeochemical behaviors in various

66 marine environments. Owing to the significantly longer half-life of  $^{210}\text{Pb}$  relative to  $^{210}\text{Po}$ , the two  
67 radionuclides are expected to be in secular equilibrium (total  $^{210}\text{Po}/^{210}\text{Pb}$  activity ratio = 1) in the  
68 ocean, assuming no net removal or addition of either radionuclide. A deficit of  $^{210}\text{Po}$  activity  
69 relative to  $^{210}\text{Pb}$  activity ( $^{210}\text{Po}/^{210}\text{Pb}$  activity ratio < 1), however, is commonly found in the upper  
70 ocean (e.g. Bacon et al., 1976; Nozaki and Tsunogai, 1976; Cochran et al., 1983; Sarin et al., 1999).  
71 This has been attributed to a higher particle reactivity of  $^{210}\text{Po}$  (higher partitioning coefficient,  $K_d$ )  
72 than  $^{210}\text{Pb}$  in seawater. Particles, therefore, become enriched in  $^{210}\text{Po}$  ( $^{210}\text{Po}/^{210}\text{Pb}$  activity ratio >  
73 1) and their sinking to deeper waters results in a  $^{210}\text{Po}$  activity deficit relative to  $^{210}\text{Pb}$  activity in  
74 the upper water column where particles are formed.

75 In this work, we describe the distributions of total and size-fractionated particulate  $^{210}\text{Po}$  and  
76  $^{210}\text{Pb}$  activity along the GEOVIDE cruise in the North Atlantic. These data are a significant  
77 contribution to the high-latitude North Atlantic  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity data set. We present a  
78 compilation of particulate  $^{210}\text{Po}/^{210}\text{Pb}$  activity ratios (AR) from previous studies in the global ocean  
79 and the results are discussed in regards to the aging of water and biochemical processes. We also  
80 describe the relationship among small particles, adsorption, and scavenging of radionuclides.  
81 These results lead to recommendations for the estimation of particulate organic carbon export flux  
82 based on the  $^{210}\text{Po}/^{210}\text{Pb}$  disequilibrium, a topic that is covered in a companion paper (Tang et al., ,  
83 companion paper submitted to this volume).

84

## 85 **2 Methods**

### 86 **2.1 Sample collection**

87 The French GEOTRACES cruise to the North Atlantic (GEOVIDE, Section GA01; May 15 –  
88 June 30, 2014) was completed on the *R/V Pourquoi Pas?*. The research vessel departed from  
89 Lisbon, Portugal, headed northwest to the Greenland shelf, crossed the Labrador Sea, and ended  
90 in St John’s, Newfoundland, Canada (Fig. 1). A rosette equipped with conductivity-temperature-  
91 depth sensors and 12 L Niskin bottles was used to collect 200 seawater samples (5 – 10 L each)  
92 from 10 full water column “super” (10 multi-cast) stations (16 – 22 depths/station) and 1 “XLarge”  
93 (5-cast) station to 800 m (station 26, 9 depths) for the determination of total  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity.  
94 Upon recovery, seawater samples were transferred to 10 L acid-cleaned containers. In addition,  
95 particulate radionuclide activities in two size classes (1-53  $\mu\text{m}$  and > 53  $\mu\text{m}$ ) were collected at 3 –  
96 10 depths per station using large volume *in-situ* filtration systems (Challenger Oceanic pumps and

97 McLane pumps) equipped with 142 mm filter holders. Each filter head contained a stacked 53  $\mu\text{m}$   
98 PETEX screen followed by a 1  $\mu\text{m}$  pore size quartz fiber QMA filter. The volume filtered was  
99 determined via flow meters mounted below each filter head, and the mean volume pumped through  
100 each head was 881 L. Once recovered, clear polyethylene caps were placed on the top of the pump  
101 heads and they were brought into a clean laboratory for sub-sampling.

102

## 103 **2.2 Total $^{210}\text{Po}$ and $^{210}\text{Pb}$**

104 Total  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities were determined from the seawater samples by the cobalt-  
105 ammonium pyrrolidine dithiocarbamate (Co-APDC) technique (Fleer and Bacon, 1984). Samples  
106 were acidified to a  $\text{pH} < 2$  with concentrated HCl immediately after collection and spiked with  
107 known amounts of  $^{209}\text{Po}$  and stable lead as chemical yield tracers. After vigorous stirring and at  
108 least 12 h of isotope equilibration, cobalt nitrate and APDC solutions were added to co-precipitate  
109 Po and Pb. Samples were filtered through a 0.45  $\mu\text{m}$  membrane filter and the filters with the  
110 precipitate were placed into clean falcon tubes, sealed with parafilm, and stored in double-bags.  
111 As the delay between sample collection and first Po plating increases, the uncertainty of the  
112 calculated  $^{210}\text{Po}$  activity also increases. In addition, it is necessary to balance counting periods with  
113 the number of samples as the uncertainty due to alpha spectrometry counting decreases by  
114 increasing the counting time. To limit the delay between sampling and processing and to ensure  
115 higher counting statistics by having more alpha spectrometers devoted to this project, sample  
116 processing and analyses were split between Universitat Autònoma de Barcelona (UAB) (samples  
117 from stations 1, 13, and 21) and Queens College (QC) (stations 26, 32, 38, 44, 60, 69, and 77).  
118 Both laboratories followed the same procedure. Briefly, the filters were digested into a solution of  
119 concentrated  $\text{HNO}_3$  and HCl, and after the solution was evaporated to dryness, the samples were  
120 recovered in 1M and 0.5 M HCl solution at UAB and QC, respectively (a 0.5-2 M HCl solution is  
121 recommended, Rigaud et al., 2013). A polished pure silver disc (Flynn, 1968) with one side  
122 covered by enamel paint was placed into the weak acid solution and heated so that the polonium  
123 nuclides were spontaneously plated onto only one side of the disc. The activities of both Po  
124 nuclides on the disc were measured by alpha spectrometry. Any  $^{210}\text{Po}$  and  $^{209}\text{Po}$  remaining in the  
125 plating solution was removed using AG 1-X8 anion exchange resin and the final solution was re-  
126 spiked with  $^{209}\text{Po}$  and stored for more than 6 months to allow ingrowth of  $^{210}\text{Po}$  from the decay of  
127  $^{210}\text{Pb}$ .

128 The  $^{210}\text{Pb}$  activity was then determined by re-plating the solutions using silver discs and  
129 measuring the ingrown  $^{210}\text{Po}$ . Two aliquots of the plating solutions for each sample were taken  
130 before the first and second platings for the measurement of total Pb concentration by inductively  
131 coupled plasma mass spectrometry (ICP-MS) to determine sample recovery during processing.  
132 The average recoveries produced by UAB and QC were  $83 \pm 11\%$  ( $n = 54$ ) and  $76 \pm 14\%$  ( $n =$   
133  $144$ ), respectively. The activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  at the time of collection were determined by a  
134 series of corrections, including nuclide decay, ingrowth, chemical recoveries, detector  
135 backgrounds, and blank contamination following the methods in Rigaud et al., (2013). The activity  
136 uncertainties from UAB were on average 8% for both  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity, while the QC  
137 uncertainties were on average 13% for  $^{210}\text{Po}$  activity and 16% for  $^{210}\text{Pb}$  activity. The greater  
138 uncertainties of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities in the samples processed at QC were due to the longer  
139 delay between sampling and first plating (68 vs. 50 d) and higher uncertainties in the determination  
140 of the recovery of lead.

141

### 142 **2.3 Particulate $^{210}\text{Po}$ and $^{210}\text{Pb}$**

143 After collection via in situ pumping, one quarter (equivalent to  $\sim 220$  L) of the PETEX screen  
144 containing  $> 53 \mu\text{m}$  or “large” particles was processed for radionuclide activity. Swimmers were  
145 carefully removed from all samples. The QMA filters containing  $1\text{-}53 \mu\text{m}$  or “small” particles  
146 were sub-sampled (2 – 4 punches of 12 mm-diameter) achieving a mean effective volume of  $\sim 66$   
147 L. The screens and punches were stored in double-bags at  $-80^\circ\text{C}$  until the analyses onshore. The  
148 particulate samples were split between the two laboratories in parallel to the seawater samples.  
149 The filters were spiked with  $^{209}\text{Po}$  tracer solution and stable lead, digested using a mixture of  
150 concentrated HF,  $\text{HNO}_3$  and HCl at UAB, but only  $\text{HNO}_3$  and HCl at QC. After multiple rounds  
151 of digestion and evaporation to near dryness, the samples were recovered in 0.5 M HCl solution.  
152 Any remaining pieces of filter which were not completely digested were carefully removed, rinsed  
153 with 0.5 M HCl solution several times, and then discarded. The analyses of the particulate  
154 radionuclide activities were identical to those for the seawater samples described in section 2.2.

155

### 156 **2.4 Concentration of suspended particulate matter (SPM)**

157 The Helene Planquette group (University of Brest, co-authors in this issue) collected  
158 subsamples from the same screens and filters that were sampled previously for radionuclides to

159 determine major phase composition (particulate organic matter (POM), lithogenic material,  
160 calcium carbonate ( $\text{CaCO}_3$ ), opal,  $\text{Fe}(\text{OH})_3$ , and  $\text{MnO}_2$ ) (references therein Lam et al., 2015). The  
161 mass concentration of SPM was calculated as the sum of the chemical dry weight of the major  
162 particulate phases.

163 The calculated SPM concentration was compared to the *in-situ* transmission data obtained from  
164 the rosette CTD sensor (Fig. S1). The overall negative relationship was statistically significant ( $R^2$   
165 = 0.7,  $n = 53$ ,  $p < 0.0001$ ), suggesting that the SPM concentrations determined were reasonable  
166 estimates of particle concentration in the water column. We used the SPM values to determine the  
167 partitioning coefficient,  $K_d$ , for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in section 4.4.

168

## 169 **2.5 Satellite-based data**

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

174

## 175 **2.6 Historical values**

176 The historical data of the particulate  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity, and the hydrological parameters  
177 (pressure, temperature, salinity, and dissolved oxygen) were obtained from databases and  
178 publications. The location, date, database address or publication name, and type of data (particulate  
179  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity or hydrological parameters) from all other studies is listed in supplemental  
180 Table S1.

181

## 182 **2.7 Apparent oxygen utilization**

183 Apparent oxygen utilization ( $\text{AOU} = \text{O}_2 \text{ saturated} - \text{O}_2 \text{ measured}$ ) is defined as the difference between  
184 the saturated oxygen at a given temperature and salinity and the measured in-situ oxygen  
185 concentration (Ito et al., 2004; Duteil et al., 2013). A positive AOU indicates either water mass  
186 aging and outgassing of oxygen or biological activity, namely respiration (e.g. Keeling et al., 1998;  
187 Boyer et al., 1999). Negative AOU, indicating that the water is oversaturated with dissolved  
188 oxygen, can appear under the conditions of an intense bloom (e.g. Coppola et al., 2017).

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

192

## 193 **2.8 Statistical analyses**

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

200

## 201 **3 Results**

### 202 **3.1 Total $^{210}\text{Po}$ and $^{210}\text{Pb}$ activities**

203 Total  $^{210}\text{Po}$  activities ( $^{210}\text{Po}_t$ ) in all samples ranged from 2.2 to 16.4 dpm 100 L<sup>-1</sup> and the mean  
204  $^{210}\text{Po}_t$  was  $8.8 \pm 2.4$  dpm 100 L<sup>-1</sup> (n = 198, Fig. 2).  $^{210}\text{Po}_t$  activities were generally low within the  
205 mixed layer and euphotic zone (15 – 47 m), slightly increased or remained relatively constant in  
206 the depth range between the mixed layer and 250 m, and then decreased with water depth at most  
207 of the stations except station 26. Near the seafloor, stations 1, 13 and 44 had a slight increase of  
208  $^{210}\text{Po}_t$  activity.

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

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

217 For all regions, significant deficits of  $^{210}\text{Po}_t$  ( $0.80 \pm 0.20$ , n = 40) were observed within the  
218 mixed layer and euphotic zone (Fig. 3). Secular equilibrium was also observed at some shallow  
219 depths (i.e. 80 m at station 44) and even in surface waters (i.e. 15 m at station 38).  $^{210}\text{Po}_t$  excesses



220 relative to  $^{210}\text{Pb}_t$ , which were larger than  $^{210}\text{Po}_t$  surface depletions at the same stations, were  
221 observed below the surface at some depths at stations 1, 13, and 21 in the West European Basin  
222 (Fig. 2). At depths below the surface to  $\sim 1500$  m in the Iceland Basin, the Irminger Sea, and the  
223 Labrador Sea, the water samples still indicated a  $^{210}\text{Po}$  deficiency (AR:  $0.84 \pm 0.17$ ,  $n = 27$ ). Secular  
224 equilibrium was generally reached near the bottom depths in all basins except at stations 13 and  
225 60 where the water samples were either enriched in  $^{210}\text{Po}_t$  ( $^{210}\text{Po}_t/^{210}\text{Pb}_t$  AR =  $1.58 \pm 0.16$ ) or  
226 depleted in  $^{210}\text{Po}_t$  ( $^{210}\text{Po}_t/^{210}\text{Pb}_t$  AR =  $0.50 \pm 0.12$ ), respectively.

227

### 228 **3.2 Particulate $^{210}\text{Po}$ and $^{210}\text{Pb}$ activities**

229 Small particulate  $^{210}\text{Po}$  ( $^{210}\text{Po}_s$ ) activities varied in a wide range from 0.08 to 4.82 dpm 100L<sup>-1</sup>  
230 (mean:  $0.76 \pm 0.63$  dpm 100L<sup>-1</sup>,  $n = 81$ ), about 83% of the values in the small particles were lower  
231 than 1.0 dpm 100L<sup>-1</sup> with higher  $^{210}\text{Po}_s$  values generally observed in the surface samples (Fig. 4,  
232 Table S2). The range of small particulate  $^{210}\text{Pb}$  ( $^{210}\text{Pb}_s$ ) activities was 0.07 to 2.89 dpm 100L<sup>-1</sup>  
233 (mean:  $0.56 \pm 0.46$  dpm 100L<sup>-1</sup>,  $n = 81$ ). The vertical profiles of  $^{210}\text{Pb}_s$  were generally similar to  
234 those of  $^{210}\text{Po}_s$ , with relatively high activity in the surface, lower activity in the subsurface and  
235 increasing activity with depth (Fig. 4). This has been seen in the North Atlantic along the  
236 GEOTRACES GA03 transect (Rigaud et al., 2015). The mean  $^{210}\text{Po}_s/^{210}\text{Pb}_s$  activity ratio (AR) was  
237  $1.43 \pm 0.96$  in the surface waters ( $n = 14$ ,  $\leq 47$  m), and  $1.57 \pm 0.90$  with all samples included ( $n =$   
238  $81$ , 8 – 3440 m). While most surface observations had an AR of  $^{210}\text{Po}_s/^{210}\text{Pb}_s$  higher than unity, 5  
239 surface samples at stations 69 and 77 showed an enrichment of  $^{210}\text{Pb}$  activity over  $^{210}\text{Po}$   
240 ( $^{210}\text{Po}_s/^{210}\text{Pb}_s$  AR:  $0.62 \pm 0.18$ ).

241 Large particulate  $^{210}\text{Po}$  ( $^{210}\text{Po}_l$ ) activities ranged from 0.01 to 0.83 dpm 100L<sup>-1</sup> with a mean of  
242  $0.10 \pm 0.12$  dpm 100L<sup>-1</sup> ( $n = 59$ , Fig. 5, Table S2). The range of  $^{210}\text{Pb}$  activity in the large particles  
243 ( $^{210}\text{Pb}_l$ ) was from 0.02 to 0.67 dpm 100L<sup>-1</sup> (mean:  $0.12 \pm 0.14$  dpm 100L<sup>-1</sup>,  $n = 59$ ). The highest  
244  $^{210}\text{Po}_l$  and  $^{210}\text{Pb}_l$  values were found at 30 m at station 26. The mean  $^{210}\text{Po}_l/^{210}\text{Pb}_l$  activity ratio (AR)  
245 was  $1.09 \pm 1.54$  in the surface waters ( $n = 14$ ,  $\leq 47$  m), and  $1.06 \pm 0.86$  when all data were  
246 considered ( $n = 59$ , 8-800 m). There were 17% of the samples with a depletion of  $^{210}\text{Po}$  activity  
247 relative to  $^{210}\text{Pb}$  activity in large particles (mean AR:  $0.49 \pm 0.23$ ), particularly in surface waters  
248 from the western section. We address this issue further in sections 4.2 and 4.3.

249 The percentages of total  $^{210}\text{Po}$  activity in the small and large particles ranged from 0.9 to 46.7%  
250 (mean:  $8.0 \pm 6.7\%$ ) and from 0.1 to 8.9% (mean:  $1.2 \pm 1.5\%$ ), respectively. The percentage of total

251  $^{210}\text{Pb}$  activity ranged from 0.7 to 21.4% (mean:  $4.9 \pm 3.8\%$ ) and from 0.2 to 5.9% (mean:  $1.1 \pm$   
252  $1.2\%$ ) in the small and large particulate phase, respectively. These values revealed that both  
253 radionuclides were predominantly present in the dissolved phase along this transect, as is  
254 commonly found in the ocean. The particulate percentages reported here are similar to the values  
255 reported from the F.S. “Meteor” cruise 32 in the North Atlantic (Bacon et al., 1976) and along the  
256 North Atlantic GA03 transect (Rigaud et al., 2015).

257 We then combined radionuclide activity on the small and large particles from the same depth  
258 as the total particulate activity. There were 56 samples in total (surface to 800 m) and 41 of them  
259 were from the upper 200 m. Most of the total particulate  $^{210}\text{Po}$  ( $^{210}\text{Po}_p$ ) and  $^{210}\text{Pb}$  ( $^{210}\text{Pb}_p$ ) activity  
260 was on the small particles, with 86% of  $^{210}\text{Po}_p$  and 80% of  $^{210}\text{Pb}_p$  on the small size fraction (data  
261 not shown). The total particulate  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  AR ( $^{210}\text{Po}_p/^{210}\text{Pb}_p$ ) had the same mean as that of  
262 the small particulate  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  AR ( $^{210}\text{Po}_s/^{210}\text{Pb}_s$ ) (Welch Two Sample t-test,  $n = 56$ ,  $p = 0.1$ ),  
263 indicating that the values of the  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  activity ratios were driven by the small particles.  
264 While the majority of particulate matter was enriched in  $^{210}\text{Po}$  ( $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR > 1), there were  
265 13 out of 56 total samples from various depths that were depleted in  $^{210}\text{Po}$  relative to  $^{210}\text{Pb}$ . The  
266  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  activity ratios from this study are compared to the results from previous studies in  
267 various oceanic regimes in section 4.2.

268

## 269 **4 Discussion**

### 270 **4.1 Total $^{210}\text{Po}$ and $^{210}\text{Pb}$ activities**

271 The overall profiles of  $^{210}\text{Po}_t$  and  $^{210}\text{Pb}_t$  activities were different among basins (Fig. 2). The  
272 deficiencies of  $^{210}\text{Po}_t$  activities with respect to  $^{210}\text{Pb}_t$  activities in the surface samples from the  
273 Iceland Basin, the Irminger Sea, and the Labrador Sea were generally greater than those from the  
274 West European Basin. Such disequilibria generally extended to the deep waters (1700 – 2950 m).  
275 In contrast,  $^{210}\text{Po}_t$  activities in the West European Basin were generally enriched relative to  $^{210}\text{Pb}_t$   
276 activities from below the surface to the bottom of the profile. In the West European Basin, the sub-  
277 surface  $^{210}\text{Po}_t$  activity excess was much larger than the surface depletion, suggesting that some  
278 external source would be needed to maintain this excess  $^{210}\text{Po}$  activity within the water column.  
279 One possible source of these sub-surface  $^{210}\text{Po}$  activity excesses below 2000 m at stations 1 and  
280 13 could be the North-East Atlantic Deep Water, lower (NEADWL) which was the dominant water  
281 mass in the Iberian Basin from 2000 m to the bottom, and had a concentration of silicate up to 48

282  $\mu\text{mol kg}^{-1}$  (García-Ibáñez et al., 2015). High activity of  $^{210}\text{Po}$  in deep samples could be due to the  
283 dissolution of diatoms or herbivore feces (Cooper, 1952). As these particles sink and dissolve,  
284  $^{210}\text{Po}$  activity may have been preferentially released to the dissolved phase compared to  $^{210}\text{Pb}$   
285 activity (Bacon et al., 1976), leading to  $^{210}\text{Po}$  excess observed in the deep waters at stations 1 and  
286 13. For the sub-surface  $^{210}\text{Po}$  activity excesses at station 1 between 400 and 1000 m where lateral  
287 inputs of particulate Fe from the margin was observed (Gourain et al., 2018), the likely process is  
288 diffusion of  $^{210}\text{Po}$  from those particles originated from the margin and such excess could be  
289 transported westwards to station 13 by lateral advection. An alternative source of  $^{210}\text{Po}$  activity  
290 excess between 50 and 250 m at stations 1 and 13 (Fig. 3) could be the eastern boundary upwelling  
291 along the coast of the Iberian Peninsula (García-Ibáñez et al., 2015). Even though no strong  
292 upwelling events were revealed from temperature and density profiles during the cruise, northerly  
293 winds favoring upwelling were recorded 2 – 3 months before the sampling (Shelley et al., 2017).  
294 The deep water may have excess  $^{210}\text{Po}$  activity due to the remineralization of sinking particles.  
295 The upwelling of this water mass prior to the sampling date could maintain such sub-surface excess  
296  $^{210}\text{Po}$  activity. Similar findings have been reported in the Cariaco Trench for the upper 300 m of  
297 the water column by Bacon et al., (1980a).

298 As atmospheric deposition is the main source of  $^{210}\text{Pb}$  to the water column (e.g. Masqué et al.,  
299 2002), we divided the GA01 transect into a western section (stn. 44 – 77) and an eastern section  
300 (stn. 1 – 38) based on atmospheric deposition boxes described in Shelley et al., (2017). Total  
301 atmospheric deposition fluxes of a suite of aerosol-sourced trace metals (TEs) were reported to be  
302 higher in the east than the west for 18 out of 19 TEs (Shelley et al., 2017). However, a two sample  
303 t-test revealed a greater mean of  $^{210}\text{Pb}_t$  activity in surface waters in the western than in the eastern  
304 section ( $p < 0.02$ , mean: 12.1 vs. 10.4 dpm 100 L<sup>-1</sup>), despite the fact that  $^{210}\text{Pb}$  is usually associated  
305 with aerosols. Even though the direct input of atmospheric  $^{210}\text{Pb}$  may be larger in the east  
306 (assuming it behaves like the other trace metals, but without aerosol  $^{210}\text{Pb}$  data we cannot confirm  
307 this), alternative inputs of  $^{210}\text{Pb}$  from freshwater (e.g., sea ice processes and meteoric water) could  
308 be a greater source of  $^{210}\text{Pb}$  activity to the west. The freshwater sources over the Greenland shelf  
309 and slope have been identified by Benetti et al., (2017), and were believed to be an important  
310 source of Fe (Tonnard et al., in review) and Al (Menzel-Barraqueta et al., in review) off of  
311 Greenland during this cruise. This result highlights the need in the future to measure  $^{210}\text{Pb}$  activity

312 simultaneously in the atmospheric and local freshwater sources in order to account for all source  
313 terms.

314

#### 315 **4.2 Total particulate $^{210}\text{Po}/^{210}\text{Pb}$ AR**

316 A proposed explanation for the depletion of  $^{210}\text{Po}$  activity relative to  $^{210}\text{Pb}$  activity ( $\text{AR} < 1$ ) in  
317 some particles is effective recycling, commonly characterized by a subsurface excess of dissolved  
318  $^{210}\text{Po}$  activity released from enriched particles leaving the surface. Bacon et al., (1976) suggested  
319 that the efficiency of this recycling could reach up to 50%, while there is no significant concurrent  
320 release of  $^{210}\text{Pb}$  activity in the water column. Laboratory studies have found the release rate of  
321  $^{210}\text{Po}$  in marine particulate matter to be significant; for example, 41% of the  $^{210}\text{Po}$  activity in  
322 euphausiid fecal pellets was released over 5 days as presented in Heyraud et al., (1976). An  
323 alternative explanation for the depletion of  $^{210}\text{Po}$  activity in particles is their lithogenic origin.  
324  $^{210}\text{Po}/^{210}\text{Pb}$  AR in lithogenic particles was reported to be similar to or less than unity (Nozaki et  
325 al., 1998; Tateda et al., 2003). In addition, the  $\text{AR} < 1$  observed at station 1 (120, 250, and 550 m)  
326 could be associated with lithogenic particles from the Iberian Margin where 100% of the  
327 particulate Fe (PFe) had a lithogenic origin while the lithogenic contribution to PFe at other  
328 stations was smaller (Gourain et al., 2018).

329 The time-series chlorophyll-a concentrations (8-day composite,  
330 <https://oceancolor.gsfc.nasa.gov>) from January to July 2014 at each station revealed bloom  
331 conditions about 4 months prior to the sampling time (Fig. 6). We estimated the days since the last  
332 bloom began prior to the sampling date for each station (Table 1) and put these data into the context  
333 of the low  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR ( $< 1$ ) in the total particles  $> 1 \mu\text{m}$  (Fig. 7). Eight stations had total  
334 particulate samples with  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR lower than unity from either shallow or deep waters.  
335 Specifically, when the time since the last bloom began was relatively short (24 – 47 d) the samples  
336 with  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR  $< 1$  were observed in the shallow waters (10 – 60 m). In contrast, as longer  
337 time (50 – 74 d) passed since the last bloom, the depths at which samples had  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR  $<$   
338 1 were found to be much deeper (120 – 500 m). The results indicated that post-bloom particles  
339 could be recycled for weeks in shallow depths and take weeks to months to sink to deeper waters.

340 The averages of  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR within the upper 200 m water column were put into a global  
341 context with previously reported results (Fig. 8). Total particulate  $^{210}\text{Po}/^{210}\text{Pb}$  AR in the open ocean  
342 in previous studies (e.g., Equatorial/western Pacific, Bellingshausen Sea, BATS, Labrador Sea)

343 were generally greater than unity. In contrast to the open ocean, the data show a distinct trend of  
344 depletion of relative  $^{210}\text{Po}$  activity in marine particles from the shallow seas of the high latitude  
345 northern hemisphere. The lowest total particulate  $^{210}\text{Po}/^{210}\text{Pb}$  AR values (Table 2, 0.4 – 0.5) were  
346 found in the Chukchi shelf (He et al., 2015) and other seas from the Eurasian sector (Barents, Kara  
347 and Laptev Seas) but also in central Arctic (Friedrich, 2011). Previous studies have observed  
348 depletion of relative  $^{210}\text{Po}$  activity in nearshore particles in the Yellow Sea (Hong et al., 1999), in  
349 the turbid waters off of western Taiwan (Wei et al., 2012), on the shelf of Woods Hole, MA  
350 (Rigaud et al., 2015), and now in the margin station off St. John's, Canada (this study). The  
351 previous authors attributed the relative depletion of particulate  $^{210}\text{Po}$  activity in the nearshore  
352 waters to the terrestrial origin/riverine input of particles with a low  $^{210}\text{Po}/^{210}\text{Pb}$  AR. This may  
353 partially explain low activity ratios in the samples from the shelf of the Arctic Ocean as well, since  
354 it receives ~ 10% of global river runoff and is the most riverine-influenced of all of the world's  
355 oceans (Opsahl et al., 1999; Carmack et al., 2006). The Arctic Basin, similarly, had widespread  
356 deficits of particulate  $^{210}\text{Po}$  activity in the upper water column during the sea-ice minimum in 2007  
357 (Roca-Marti et al., in review). Besides shelf particles, the authors suggest that other particle types  
358 could also play a role in lowering the particulate AR, including sea-ice sediments, remineralized  
359 material, fecal pellets, and picoplankton aggregates.

360

#### 361 **4.3 Relationship between total particulate $^{210}\text{Po}/^{210}\text{Pb}$ AR and AOU**

362 AOU is a time-integrated measure of the amount of oxygen removed during the  
363 biogeochemical processes (e.g. respiration, remineralization, oxidation) in the ocean interior.  
364 Therefore, AOU is a product of apparent oxygen utilization rate (AOUR) and the age of water  
365 mass (e.g. Stanley et al., 2012), i.e. high AOU could be due to either intense biogeochemical  
366 processes that have occurred in a short period of time (young water mass) or weaker processes  
367 over a longer period of time (old water mass). Consequently, the rate of these biogeochemical  
368 processes and time (water mass age) would have different/similar impacts on the  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR  
369 value depending on the initial AR in the particles and the natural of the particles. For example, the  
370  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR would tend to increase with time if the initial AR is  $< 1$  because particulate  $^{210}\text{Po}$   
371 activity would increase from the decay of  $^{210}\text{Pb}$  and trend towards secular equilibrium  
372 ( $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR = 1), and to decrease with time if the initial AR is  $> 1$  as the original excess of  
373 particulate  $^{210}\text{Po}$  activity would disappear after 7 half-lives of  $^{210}\text{Po}$ . In contrast, oxygen

374 consumption due to bacterial remineralization would preferentially release  $^{210}\text{Po}$  activity from  
375 particles into the dissolved pool (e.g. Stewart et al., 2008), leading to a lower  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR in  
376 those particles.

377 The combination of average  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR and their corresponding average AOU in the  
378 upper 200 m at 40 stations from 4 independent studies, including ARK-XXII/2 (77.38 – 87.83 °N,  
379  $n = 15$ ) in the Arctic, BOFS (48.89 – 49.87 °N,  $n = 7$ ), GA03 (22.38 – 39.70 °N,  $n = 7$ ), and GA01  
380 (this study, 40.33 – 59.80 °N,  $n = 11$ ) in the North Atlantic (see map in Fig. 8) suggests two distinct  
381 linear trends (Fig. 9). When AOU was lower than  $25 \mu\text{mol kg}^{-1}$ , the  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR was found to  
382 be greater than unity, together with a linear negative relationship ( $n = 27$ ,  $R^2 = 0.5$ ,  $p < 0.001$ )  
383 towards the AOU at  $25 \mu\text{mol kg}^{-1}$ . In contrast, AOU values greater than  $25 \mu\text{mol kg}^{-1}$  were  
384 coincident with a  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR  $< 1$ , and there was a linear positive relationship ( $n = 12$ ,  $R^2 =$   
385  $0.4$ ,  $p = 0.03$ ) towards the highest AOU values measured. The two contradictory linear trends likely  
386 reflect the nature of the particles. For example, the observation of  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR  $> 1$  with AOU  
387  $< 25 \mu\text{mol kg}^{-1}$  may suggest relatively fresh/organic particles in the young water mass. When AOU  
388 increases either due to water mass aging or higher AOUR, the  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR decreases with a  
389 slope of  $-0.17 \pm 0.04$ . On the other hand, refractory/lithogenic particles may be suggested by the  
390 observation of  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR  $< 1$  with AOU  $> 25 \mu\text{mol kg}^{-1}$ . For those particles, increasing in  
391 AOU either due to water mass aging or higher AOUR would change the  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR to a  
392 much lesser degree than that for organic particles with a slope of  $0.008 \pm 0.003$ . This explanation,  
393 however, appears to only hold for the high latitude Northern Hemisphere where  $^{210}\text{Po}_p/^{210}\text{Pb}_p$   
394 activity ratios were generally lower than those in the other oceanic settings (Fig. 8). In the high  
395 latitude Southern Hemisphere near Antarctic (e.g., ANT-X/6), for example, there is no apparent  
396 relationship between  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  activity ratios and AOU. This relationship (or lack thereof)  
397 deserves more study in the future.

398

#### 399 **4.4 Relationship among small particles, adsorption, and scavenging**

400 The partitioning coefficient,  $K_d$  ( $\text{L kg}^{-1}$ ), has been used to describe the particle adsorption  
401 behavior of radionuclides. It is defined as the ratio of the adsorbed radionuclide activity ( $A_p$ , dpm  
402  $100\text{L}^{-1}$ ) to the dissolved radionuclide activity ( $A_d$ , dpm  $100\text{L}^{-1}$ ), normalized by the suspended  
403 particulate matter concentration ( $SPM$ ,  $\mu\text{g L}^{-1}$ ):

404 
$$K_d = \frac{A_p}{A_d} \times \frac{1}{SPM} 10^9 \quad (1)$$

405 Owing to the different biological and chemical behaviors of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , the interpretation  
406 of measured  $K_d$  for  $^{210}\text{Po}$  ( $K_d(\text{Po})$ ) may not be as clear as that for  $^{210}\text{Pb}$  ( $K_d(\text{Pb})$ ). As claimed  
407 previously in Tang et al., (2017),  $K_d(\text{Po})$  is complicated because it appears to reflect both the  
408 surface adsorption and potential bioaccumulation.

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

420 The average values of  $K_d(\text{Po})$  was 1.6 times of those of  $K_d(\text{Pb})$  in both small and total  
421 particulate phases, suggesting a higher affinity with particles for  $^{210}\text{Po}$  with respect to  $^{210}\text{Pb}$ , which  
422 is commonly observed in the global ocean (Bacon et al., 1988; Hong et al., 1999; Masqué et al.,  
423 2002; Wei et al., 2014; Tang et al., 2017). The  $K_d$  values for the small particulate phase were  
424 slightly higher than those for the total particulate phase but overall these values were very similar  
425 for both radionuclides (Fig. 10), suggesting that adsorption/scavenging of radionuclides was driven  
426 by small particles along the transect. In addition, there are increasing studies which argue that  
427 small particles can form aggregates that sink, and their contribution to carbon export could be  
428 larger than previously thought (e.g. Richardson and Jackson, 2007; Lomas and Moran, 2011;  
429 Amacher et al., 2013; Puigcorbé et al., 2015). We, therefore, recommend combining the activities  
430 of both small and large particles into a total particulate fraction in order to explain total  $^{210}\text{Po}/^{210}\text{Pb}$   
431 disequilibria in the surface waters, and utilizing the characteristics of the total particles (instead of  
432 just the large particles) in the estimation of the POC export fluxes (Tang et al., companion paper  
433 submitted to this volume).

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

444

## 445 **5 Conclusions**

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

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

463 Over 80% of the particulate radionuclide activity was on small particles, indicating that the  
464 scavenging of both radionuclides was driven by small particles. Therefore, we suggest considering



465 the activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  from both small and large particles in order to study the water  
466 column  $^{210}\text{Po}/^{210}\text{Pb}$  disequilibria and quantify POC export along the GA01 transect. This has been  
467 addressed in a companion paper in this issue. We recommend that both small and large particles  
468 should be sampled for POC/ $^{210}\text{Po}$  estimates for the application of the  $^{210}\text{Po}/^{210}\text{Pb}$  method in future  
469 studies of POC export.

470  
471

## 472 **Acknowledgements**

473

474 Thank you to the chief scientists (G. Sarthou and P. Lherminier) of the GEOVIDE cruise, and the  
475 captain (G. Ferrand), and crew of the *R/V Pourquoi Pas?* for their support of this work. Many  
476 thanks to P. Branellec, F. Desprez de Gésincourt, M. Hamon, C. Kermabon, P. Le Bot, S. Leizour,  
477 O. Ménage, F. Pérault, and E. de Saint-Léger for their technical support during the GEOVIDE  
478 expedition, and to C. Schmechtig for the GEOVIDE database management. P. Lam is also  
479 acknowledged for providing two modified McLane ISP. Special thanks go to the member of the  
480 pump group including F. Planchon, V. Sanial, and C. Jeandel. The author would like to thank C.  
481 Mariez, S. Roig, F. Planchon, and H. Planquette who helped in providing particle composition  
482 data. We also would like to acknowledge the funding agencies: the French National Research  
483 Agency (ANR-13-BS06-0014, ANR-12-PDOC-0025-01), the French National Center for  
484 Scientific Research (CNRS-LEFE-CYBER), the LabexMER (anr-10-LABX-19), and Ifremer.  
485 Funding was provided to P. Masque by the Generalitat de Catalunya (Grant 2017 SGR-1588). This  
486 work contributes to the ICTA ‘Unit of Excellence’ (MinECo, MDM2015-0552). G. Stewart and  
487 Y. Tang were supported by NSF award #OCE 1237108. M. Castrillejo and M. Roca-Martí were  
488 funded by an FPU PhD studentship (AP-2012-2901 and AP2010-2510, respectively) from the  
489 Ministerio de Educación, Cultura y Deporte of Spain. M. Castrillejo was also supported by the  
490 ETH Zurich Postdoctoral Fellowship Program (17-2 FEL-30), co-funded by the Marie Curie  
491 Actions for People COFUND Program. Additional thanks go to G. Hemming (Queens College)  
492 and T. Rasbury (Stony Brook University) for laboratory assistance with the ICP-MS analyses. We  
493 also thank two anonymous reviewers for their constructive comments to improve the manuscript.

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Table 1. Biological characteristics of the water column determined by chlorophyll-a concentration (8-day composite) from 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  $^{210}\text{Po}_p/^{210}\text{Pb}_p < 1$  and their corresponding depths are also shown. *NA* indicates that all samples from the corresponding depth range had  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  equal to or greater than 1 (no sample with  $^{210}\text{Po}_p/^{210}\text{Pb}_p < 1$ ).

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

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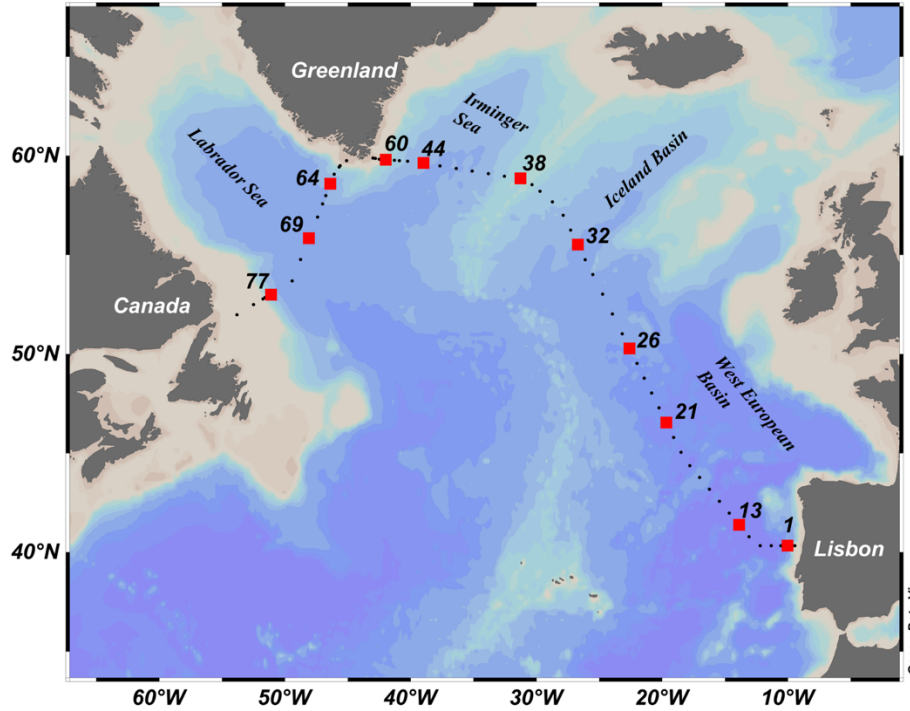
813 Table 2. The compilation of total particulate  $^{210}\text{Po}/^{210}\text{Pb}$  activity ratios ( $^{210}\text{Po}_p/^{210}\text{Pb}_p$ ) averaged in the upper 200 m, including this  
 814 study.

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

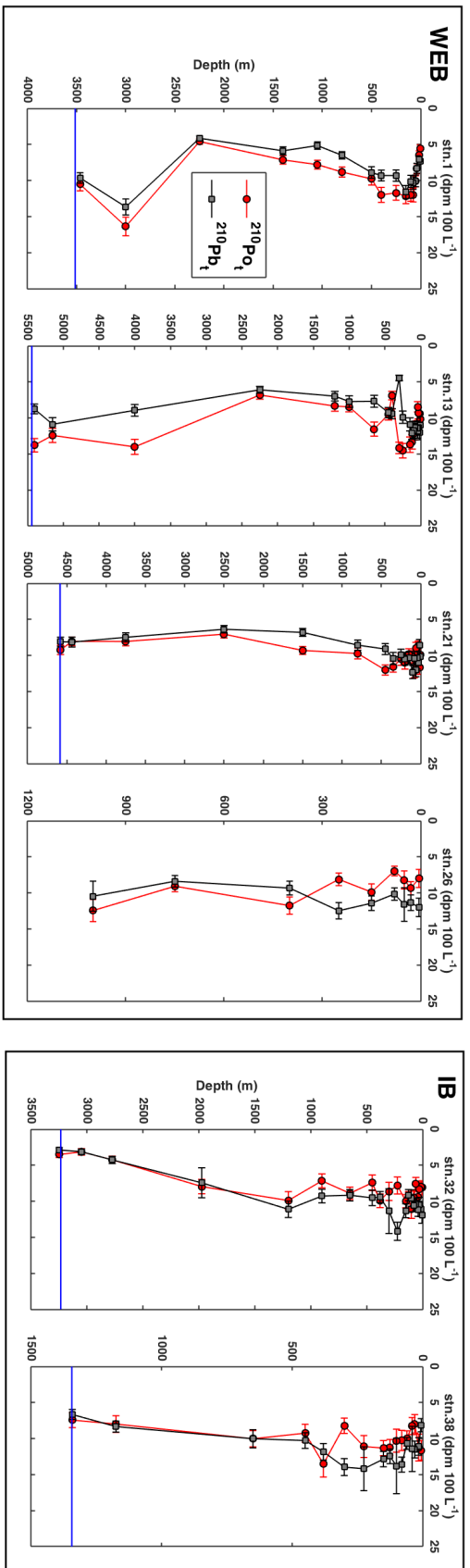
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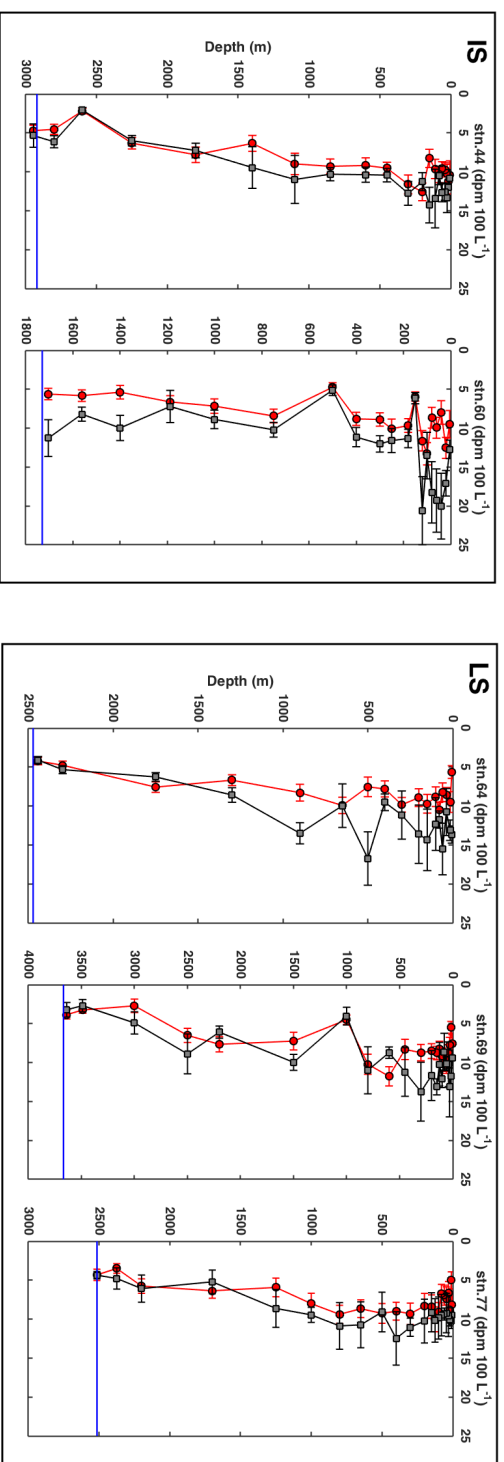


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818 Fig. 1. Map of the GEOVIDE cruise track (black dots) and the 11 stations sampled for  $^{210}\text{Po}$  and  
 819  $^{210}\text{Pb}$  activity (red squares). Each sampling location is labeled with a station number. The  
 820 sampling stations are divided into 4 regions (from east to west): West European Basin (stations  
 821 1, 13, 21, 26), Iceland Basin (stations 32, 38), Irminger Sea (stations 44, 60), and Labrador Sea  
 822 (stations 64, 69, 77).



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Fig. 2. The depth profiles of total  $^{210}\text{Po}$  ( $^{210}\text{Po}_t$ , red circles) and  $^{210}\text{Pb}$  activities ( $^{210}\text{Pb}_t$ , grey squares) along GEOVIDE section. The

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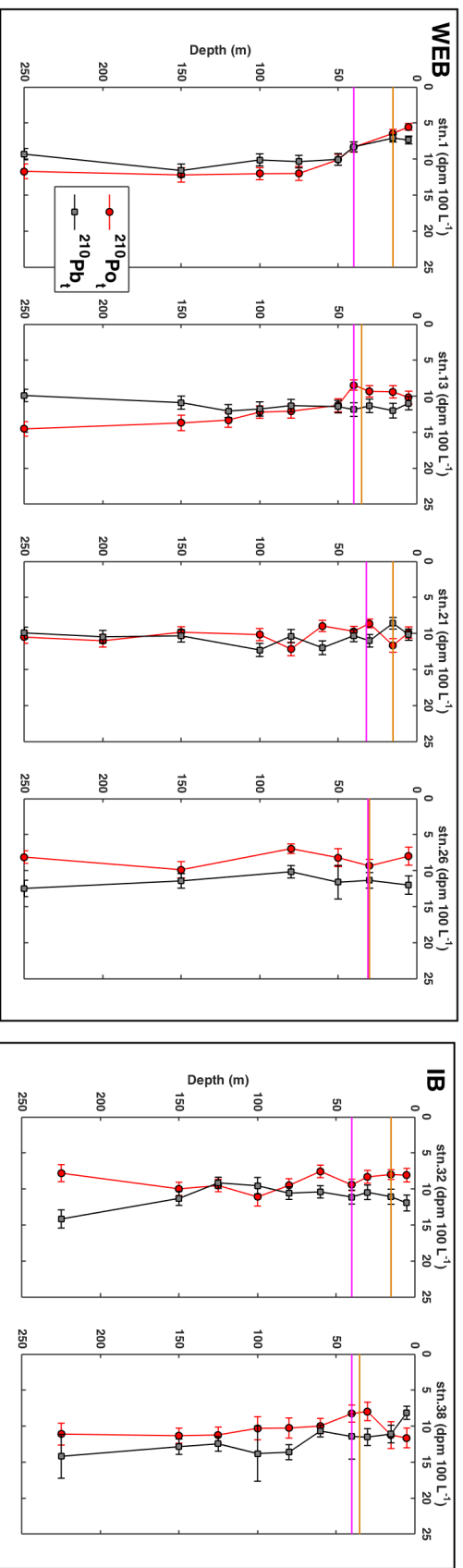
horizontal blue line is the bottom depth, which coincided with the deepest water sample except for station 26 which was sampled only

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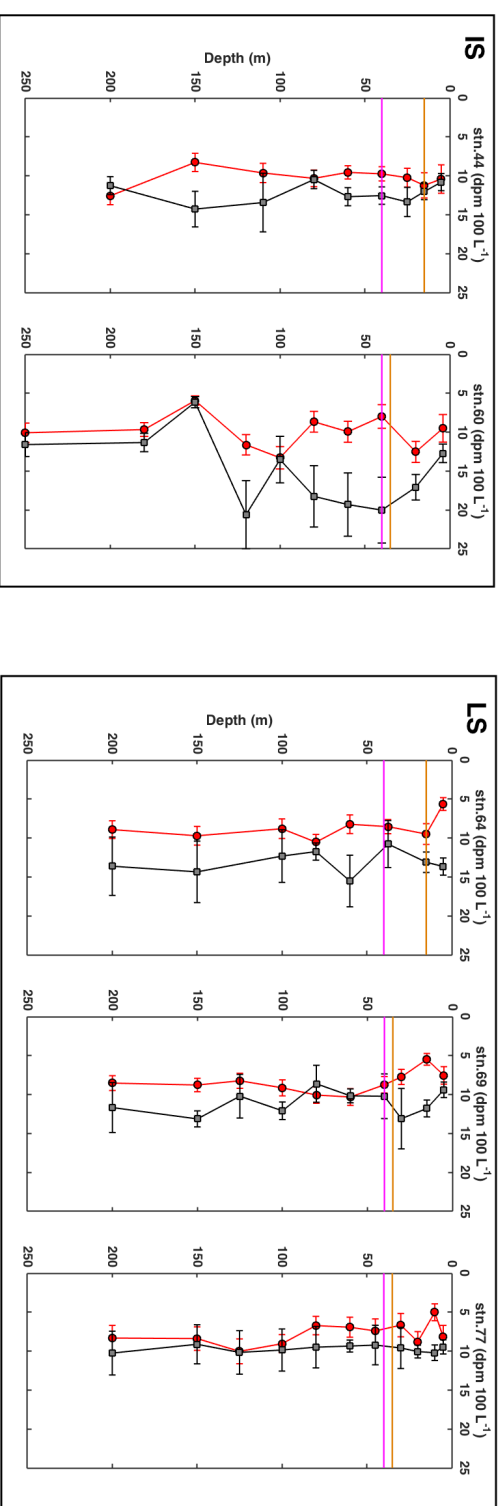
down to 1000 m. Note that the depth scale for each plot may be different. The profiles are shown in the order of sampling date with

828 the region indicated on the top left of each box: West European Basin (WEB), Iceland Basin (IB), Irminger Sea (IS), Labrador Sea  
829 (LS).

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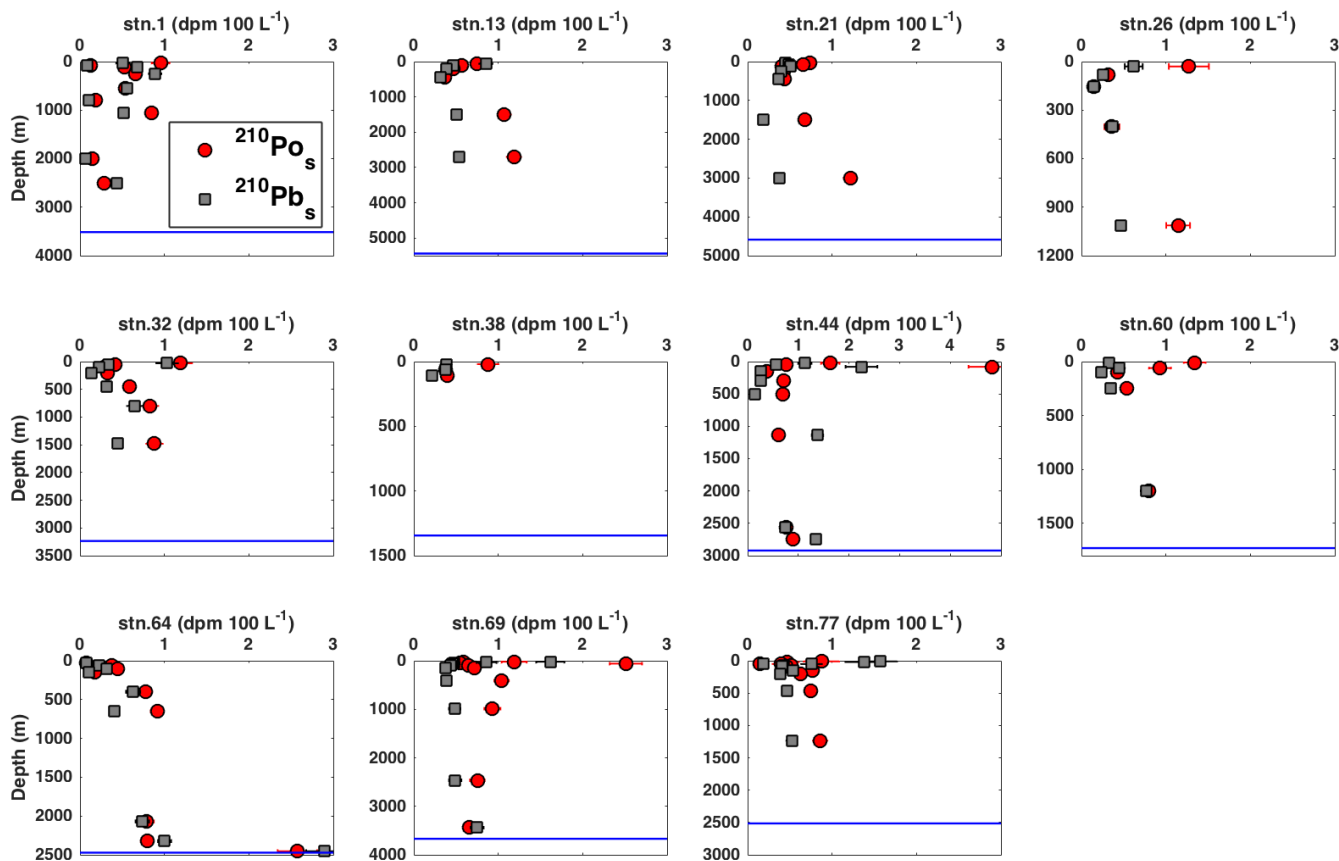


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833 Fig. 3. The upper 250 m of the depth profiles of total  $^{210}\text{Po}$  ( $^{210}\text{Po}_t$ , red circles) and  $^{210}\text{Pb}$  activities ( $^{210}\text{Pb}_t$ , grey squares) along the  
 834 GEOVIDE section. The horizontal orange and magenta lines denote the mixed layer depth (MLD) and the base of the euphotic zone

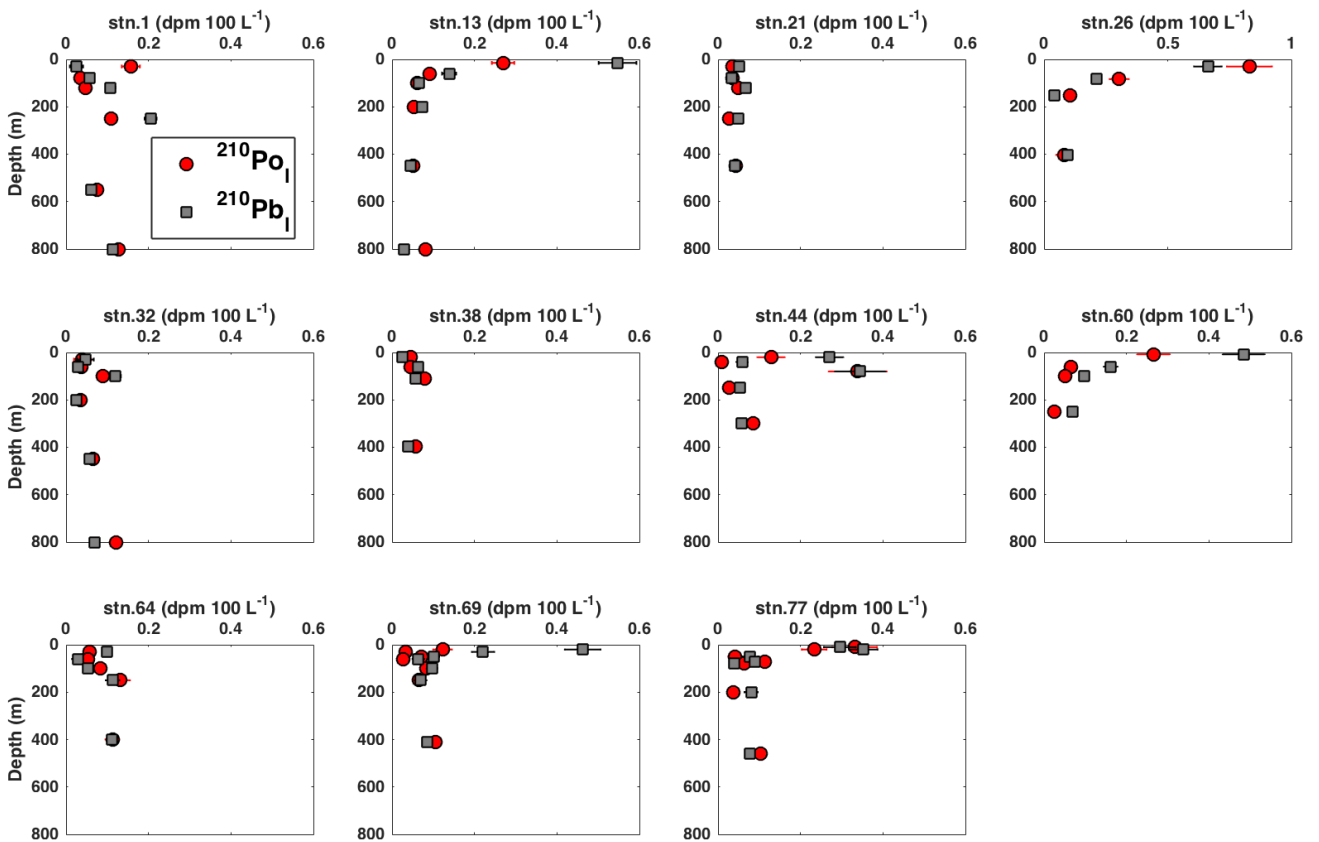
835 (Z<sub>1</sub>%), respectively. The depth profiles are shown in the order of sampling and grouped by region (refer to Fig. 2 for the text  
836 abbreviations).





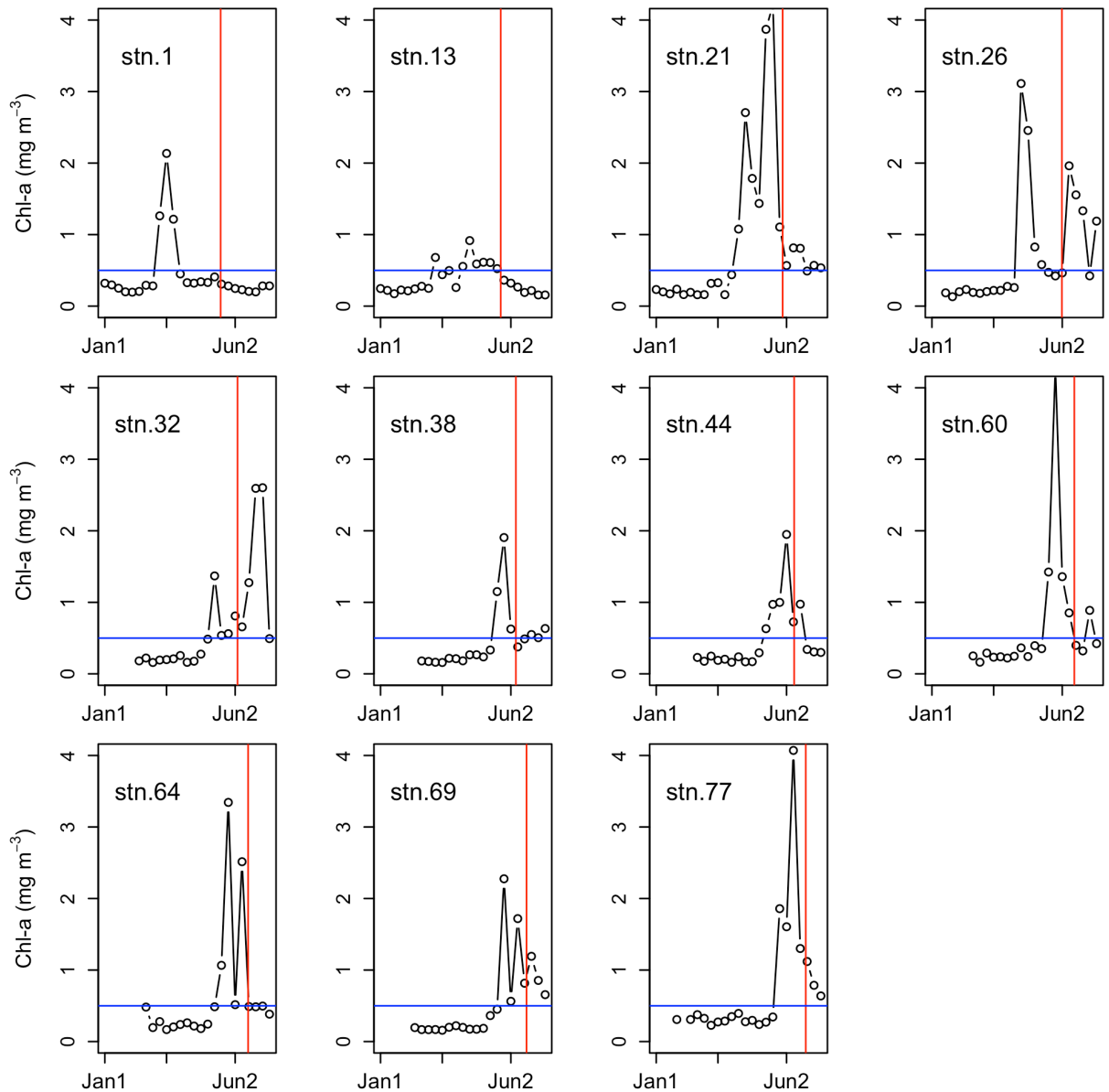
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838 Fig. 4. Vertical profiles of the particulate  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity in the small size fraction (1-53  
 839  $\mu\text{m}$ ,  $^{210}\text{Po}_s$ ,  $^{210}\text{Pb}_s$ ). Note that the depth scale may differ among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



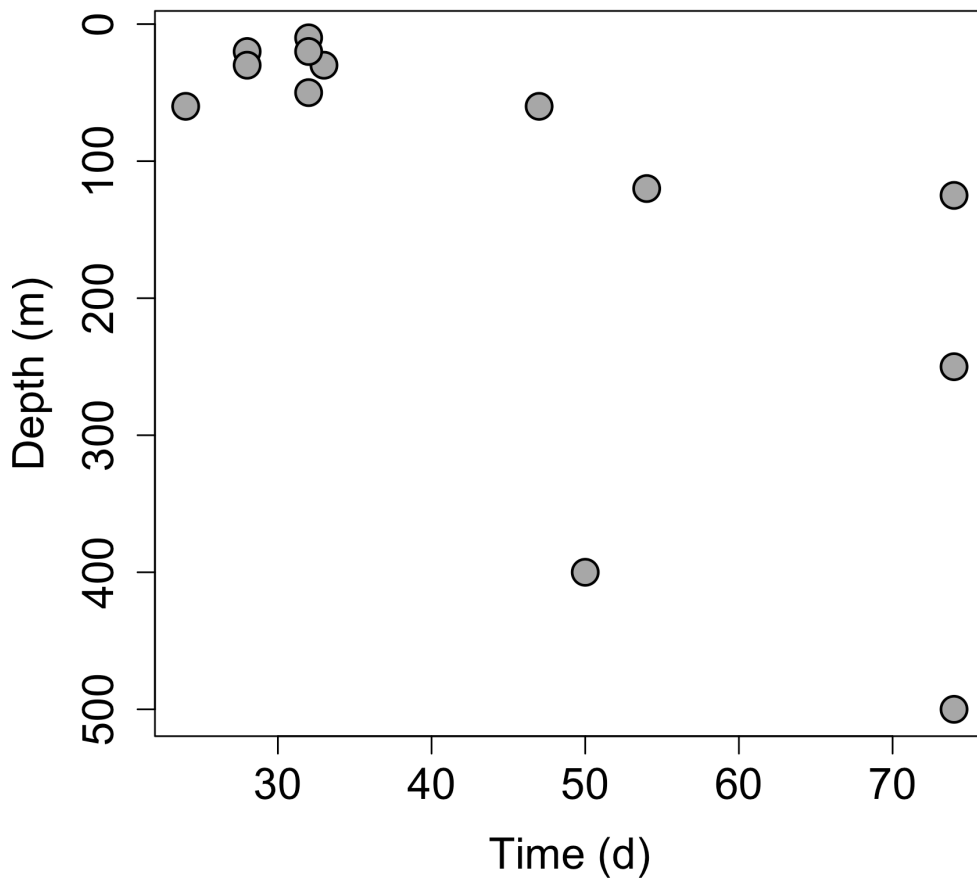
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843 Fig. 5. The vertical profiles of the particulate  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity in the large size fraction (>  
 844  $53 \mu\text{m}$ ,  $^{210}\text{Po}_I$ ,  $^{210}\text{Pb}_I$ ) in the top 800 m. Note that the activity scale at Station 26 differs from the  
 845 scale on all other plots.



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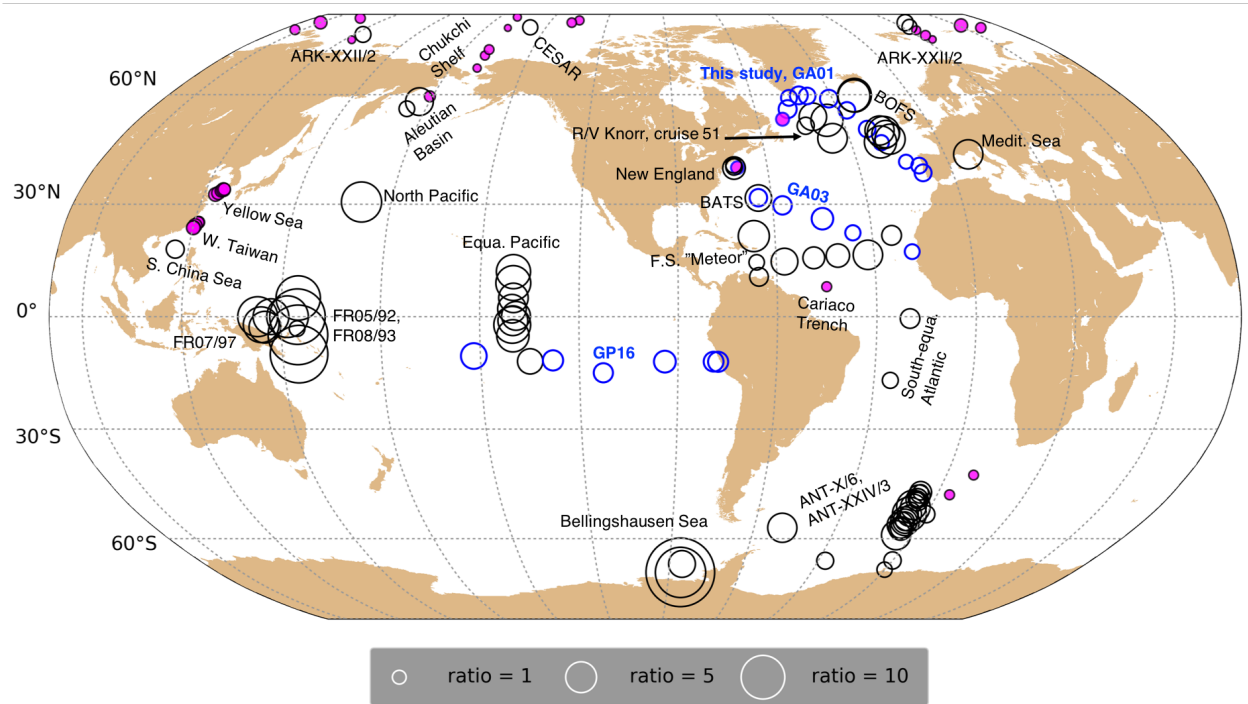
Fig. 6. Time-series (January 1 – July 12, 2014) chlorophyll-a concentrations (8-day averages) from Aqua MODIS (<https://oceancolor.gsfc.nasa.gov>) at each station along the GA01 transect. The vertical red line denotes the sampling date at each station. The horizontal blue line denotes chlorophyll-a concentration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



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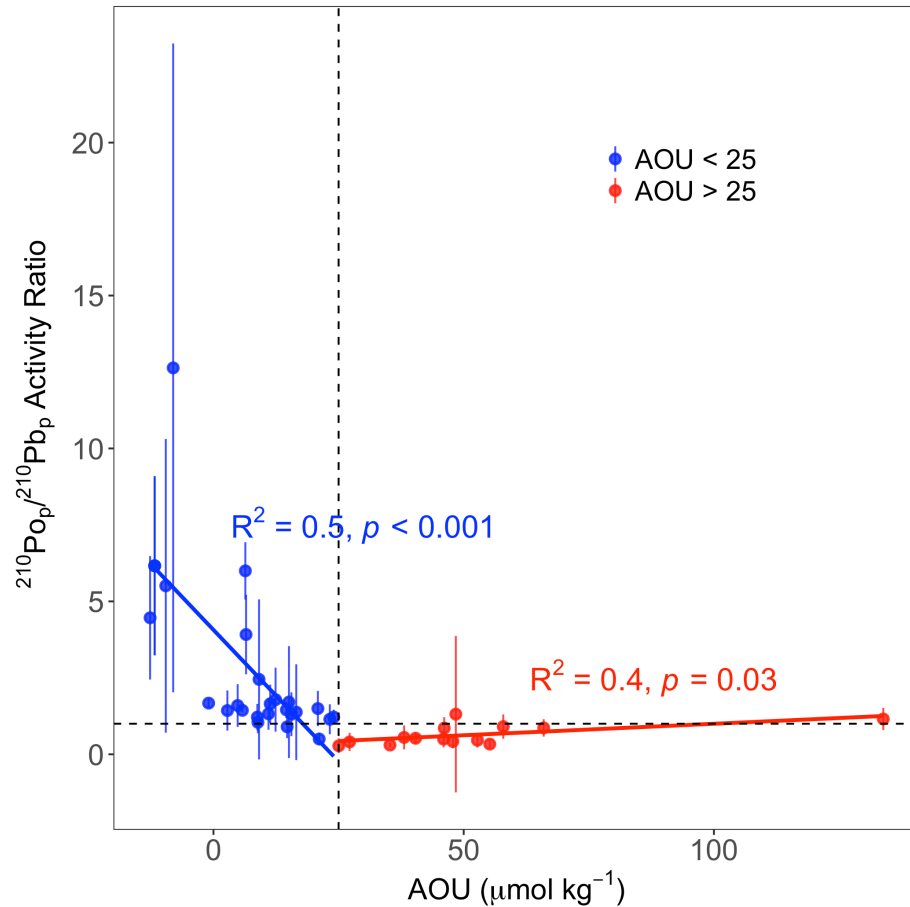
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855 Fig.7. Depths at which the total particulate ( $> 1 \mu\text{m}$ )  $^{210}\text{Po}/^{210}\text{Pb}$  activity ratio was lower than  
 856 unity vs. the time since the last bloom (data is presented in Table 1).



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Fig. 8. Comparison of particulate  $^{210}\text{Po}/^{210}\text{Pb}$  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 in the Stewart lab 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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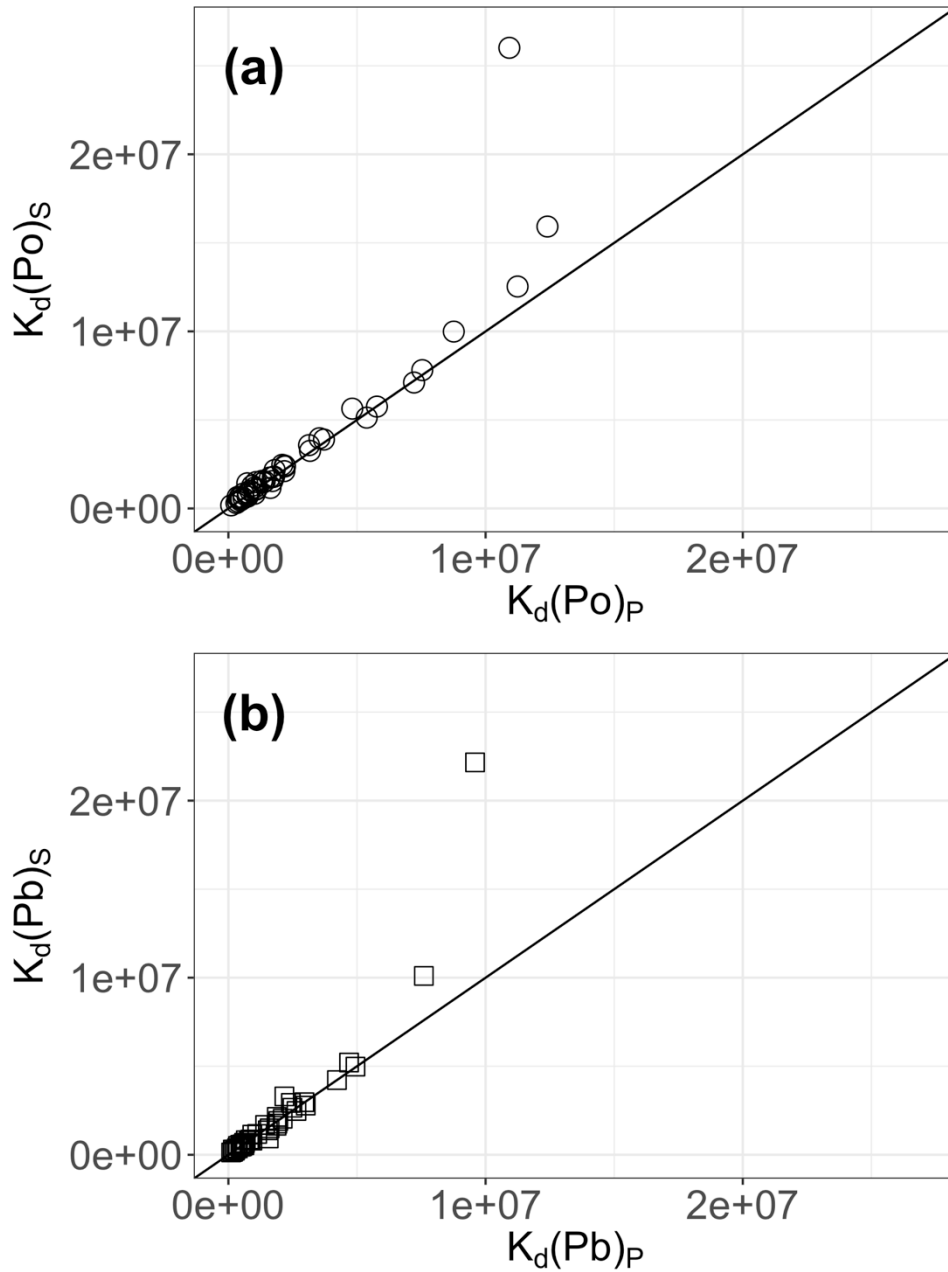
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Fig. 9. The relationship between AOU ( $\mu\text{mol kg}^{-1}$ ) and total particulate  $^{210}\text{Po}/^{210}\text{Pb}$  activity ratio ( $^{210}\text{Po}_p/^{210}\text{Pb}_p$ ) from the upper 200 m in the northern hemisphere ( $> 22^\circ\text{N}$ ) investigated by a linear regression model (red and blue lines). The 40 stations include data from previous studies, ARK-XXII/2 ( $77.38\text{-}87.83^\circ\text{N}$ ,  $n = 15$ ) in the Arctic, BOFS ( $48.89\text{-}49.87^\circ\text{N}$ ,  $n = 7$ ), GA03 ( $22.38\text{-}39.70^\circ\text{N}$ ,  $n = 7$ ), and this study, GA01 ( $40.33\text{-}59.80^\circ\text{N}$ ,  $n = 11$ ) in the North Atlantic. The horizontal dashed line represents  $^{210}\text{Po}_p/^{210}\text{Pb}_p$  AR = 1 and the vertical dashed line represents  $\text{AOU} = 25 \mu\text{mol kg}^{-1}$ . Blue circles denote  $\text{AOU} < 25 \mu\text{mol kg}^{-1}$ , while red circles  $\text{AOU} > 25 \mu\text{mol kg}^{-1}$ .



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