

1 **^{230}Th and ^{231}Pa on GEOTRACES GA03, the U.S. GEOTRACES North Atlantic Transect,**
2 **and implications for modern and paleoceanographic chemical fluxes**

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27

28 **Abstract**

29

30 The long-lived uranium decay products ^{230}Th and ^{231}Pa are widely used as quantitative tracers of
31 adsorption to sinking particles (scavenging) in the ocean by exploiting the principles of radioactive
32 disequilibria. Because of their preservation in the Pleistocene sediment record and through largely
33 untested assumptions about their chemical behavior in the water column, the two radionuclides
34 have also been used as proxies for a variety of chemical fluxes in the past ocean. This includes the
35 vertical flux of particulate matter to the seafloor, the lateral flux of insoluble elements to
36 continental margins (boundary scavenging), and the southward flux of water out of the deep North
37 Atlantic. In a section of unprecedented vertical and zonal resolution, the distributions of ^{230}Th and
38 ^{231}Pa across the North Atlantic shed light on the marine cycling of these radionuclides and further
39 inform their use as tracers of chemical flux. Enhanced scavenging intensities are observed in
40 benthic layers of resuspended sediments on the eastern and western margins and in a hydrothermal
41 plume emanating from the Mid-Atlantic Ridge. Boundary scavenging is clearly expressed in the
42 water column along a transect between Mauritania and Cape Verde which is used to quantify a
43 bias in sediment fluxes calculated using ^{230}Th -normalization and to demonstrate enhanced ^{231}Pa
44 removal from the deep North Atlantic by this mechanism. The influence of deep ocean ventilation
45 that leads to the southward export of ^{231}Pa is apparent. The $^{231}\text{Pa}/^{230}\text{Th}$ ratio, however,
46 predominantly reflects spatial variability in scavenging intensity, complicating its applicability as
47 a proxy for the Atlantic meridional overturning circulation.

48

49 **1. Introduction**

50 The motivations to quantify chemical fluxes in the ocean are manifold. For instance, marine
51 biological productivity is set by the balance between nutrient sources and sinks in surface waters
52 and global climate is influenced by the redistribution of heat and salt associated with the ocean's
53 overturning circulation. The well-known rates of radioactive production and decay of ^{230}Th and
54 ^{231}Pa (half-lives 75.69 kyr (Cheng et al., 2000) and 32.76 kyr (Robert et al., 1969), respectively),
55 in addition to their insoluble nature, make them attractive tools to quantify the rates of the marine
56 processes in which they are involved. These include removal from the water column by adsorption
57 to particles (scavenging, related to biological productivity), redistribution by ocean circulation
58 (related to heat transport), and sedimentation to the seafloor (providing a record of past biological
59 productivity, ocean circulation, and more). Unfortunately, the influences of these processes on
60 radionuclide distributions are potentially convolved. This study aims to utilize the spatial
61 distribution of ^{230}Th and ^{231}Pa across the U.S. GEOTRACES North Atlantic Transect (Fig. 1) to
62 characterize the modern cycling of these isotopes in an effort to more completely calibrate their
63 use as flux tracers in the modern and past ocean.

64 Because their production (^{234}U and ^{235}U decay, respectively) is uniform throughout the
65 ocean (Andersen et al., 2010; Delanghe et al., 2002; Robinson et al., 2004; Weyer et al., 2008), the
66 key question in ^{230}Th and ^{231}Pa cycling in the water column is the balance between removal
67 mechanisms. These are primarily (1) the downward flux by scavenging onto sinking particles and
68 (2) lateral fluxes by advection and eddy diffusion. If lateral fluxes can be neglected, the
69 concentration of the scavenged nuclide is expected to increase linearly with depth, representing an
70 “equilibrium” between adsorption onto, and desorption from, vertically homogeneous sinking

71 particles, a concept known as reversible scavenging (Bacon and Anderson, 1982; Krishnaswami
72 et al., 1976; Nozaki et al., 1981).

73 Deviations from linearity in the radionuclide profiles therefore signal where this vertical
74 equilibrium is perturbed by lateral fluxes or where the scavenging intensity has changed. This is
75 admittedly a simple approach, as relatively linear depth profiles are not inconsistent with some
76 lateral flux by dispersion (Roy-Barman, 2009; Venchiarutti et al., 2008). In a basin-scale view,
77 nonetheless, characterizing anomalies to the predictions of reversible scavenging is our first step
78 in deconvolving the oceanic ^{230}Th and ^{231}Pa cycles. Three such anomalies, boundary scavenging,
79 the effects of recently ventilated deep water, and bottom scavenging, appear in unprecedented
80 detail in our North Atlantic section (Fig. 1). We now provide a context for these findings.

81 *1.1 Boundary scavenging of ^{230}Th and ^{231}Pa*

82 Boundary scavenging (Bacon, 1988; Bacon et al., 1976; Spencer et al., 1981) is the
83 enhanced removal of scavenged-type elements (Bruland and Lohan, 2003) at ocean margins. When
84 lateral gradients in particle flux exist, as between biologically productive ocean margin regions
85 and oligotrophic ocean interior regions, insoluble elements are removed from the water column by
86 scavenging to a greater extent at the margin versus the interior. The resulting gradient in
87 radionuclide concentration produces a dispersive flux toward the margin from the interior. Lateral
88 transport in the water column toward ocean margins is more significant for ^{231}Pa than for ^{230}Th
89 because it is more slowly removed downward by scavenging. The residence time with respect to
90 scavenging of ^{231}Pa is 50-200 yrs while that for ^{230}Th is 10-40 yrs (Henderson and Anderson,
91 2003). On the basis of the boundary scavenging concept alone, elevated $^{231}\text{Pa}/^{230}\text{Th}$ ratios in both
92 the dissolved and particulate phase at ocean margins are expected (Fig. 2). Prior to this study, the
93 lateral gradients in the dissolved $^{231}\text{Pa}/^{230}\text{Th}$ ratio or in dissolved ^{230}Th (^{231}Pa) concentrations,

94 predicted by the boundary scavenging concept, have not been definitively observed in the North
95 Atlantic.

96 Modeling efforts have concluded that in ~70% of the ocean, ^{230}Th is redistributed laterally
97 by no more than 30% of its in situ production in the water column (Henderson et al., 1999),
98 consistent with available observations from sediment traps (Yu et al., 2001). However, on the basis
99 of sedimentary records some authors have argued that water column ^{230}Th redistribution could be
100 much greater than 30% due to boundary scavenging-type mechanisms, specifically along the
101 equator in the Pacific (Broecker, 2008; Lyle et al., 2005; Lyle et al., 2007). This claim derives
102 from a concern regarding ^{230}Th -normalization, a method for calculating sediment accumulation
103 rates on the basis of sedimentary ^{230}Th concentrations (Bacon, 1984; François et al., 2004). This
104 method assumes that the burial flux of ^{230}Th is equal to its rate of production by ^{234}U decay in the
105 overlying water column, which allows one to correct for the lateral redistribution of sediments at
106 the seafloor (sediment focusing). Because glacial-interglacial changes in sediment focusing have
107 enhanced or diminished apparent accumulation rates by more than a factor of 2 (François et al.,
108 1990; Suman and Bacon, 1989), the approach has been defended on the basis that neglecting a
109 relatively small bias in the assumption that ^{230}Th burial is equivalent to its production in the
110 overlying water column is justified (François et al., 2007; Siddall et al., 2008). One aim of this
111 study is to quantitatively estimate the magnitude of ^{230}Th redistribution due to boundary
112 scavenging.

113 While the effect of boundary scavenging of ^{231}Pa is well-expressed in the Pacific (Anderson
114 et al., 1983; Anderson et al., 1990; Walter et al., 1999; Yang et al., 1986), it is considered to be
115 suppressed in the Atlantic. This is because this basin is ventilated by southward flowing North
116 Atlantic Deep Water (NADW) on timescales (<100-200 yrs) (Broecker et al., 1991) shorter than

117 the Pa residence time with respect to scavenging (Walter et al., 1999; Yu et al., 1996; Yu et al.,
118 2001). This means Pa can be transported south by deep water flow before it can be dispersed to
119 North Atlantic margins. Although some studies have found evidence, in the form of sedimentary
120 $^{231}\text{Pa}/^{230}\text{Th}$ activity ratios above that produced in seawater by uranium decay of 0.093, for the
121 enhanced removal of ^{231}Pa in the upwelling area off Northwest Africa (Legeleux et al., 1995;
122 Lippold et al., 2012b; Mangini and Diester-Haas, 1983), studies of the North American (Anderson
123 et al., 1994; Lippold et al., 2012a) and the northern Brazil (Lippold et al., 2011) margins do not
124 support boundary scavenging of Pa. The dissolved $^{231}\text{Pa}/^{230}\text{Th}$ distribution toward the margins of
125 our transect (Fig. 1) will be used to determine the significance of boundary scavenging in the North
126 Atlantic in light of its recent ventilation.

127 *1.2 The impact of Atlantic circulation*

128 The possibility of boundary scavenging notwithstanding, previous studies have
129 demonstrated that deepwater distributions of ^{230}Th and ^{231}Pa are significantly perturbed by the
130 influence of the recent ventilation of NADW (Luo et al., 2010; Moran et al., 1997; Moran et al.,
131 1995; Moran et al., 2002; Scholten et al., 2001; Vogler et al., 1998). Deep convection at sites of
132 deep water formation results in the injection to depth and propagation along deepwater flow paths
133 of ^{231}Pa and ^{230}Th concentrations which are lower than predicted by reversible scavenging (Moran
134 et al., 1997; Moran et al., 1995; Moran et al., 2002). As the water mass ages, isolated from further
135 perturbations to scavenging equilibrium, dissolved ^{230}Th concentrations increase due to exchange
136 with sinking particles, reaching a steady-state distribution relatively rapidly (determined by the
137 residence time of 10-40 yrs), while ^{231}Pa responds more slowly (residence time of 50-200 yrs)
138 because of the differing scavenging rates of the two elements (Moran et al., 2001; Rutgers v. d.
139 Loeff and Berger, 1993). The longer residence time of ^{231}Pa allows for its southward export with

140 NADW, leaving a ^{231}Pa deficit in deep North Atlantic sediments (Yu et al., 1996). This is the basis
141 for using the sedimentary $^{231}\text{Pa}/^{230}\text{Th}$ ratio as an indicator of the strength of the Atlantic meridional
142 overturning circulation (McManus et al., 2004). The present water column transect is also intended
143 to document the impact of ventilation on ^{231}Pa and the $^{231}\text{Pa}/^{230}\text{Th}$ ratio.

144 In the absence of variations in scavenging intensity, one expects ^{230}Th and ^{231}Pa
145 concentrations and the $^{231}\text{Pa}/^{230}\text{Th}$ ratio to increase with water mass age or time since deep water
146 formation. The strongest response to ageing occurs within 1 to 2 water column residence times
147 after deep water formation. Our section is appropriate to test this prediction because deep water
148 age, or the time since deep-water (as averaged below 2 km) has been isolated from the atmosphere,
149 ranges from <50 yrs in the west to >250 yrs in the east (Broecker et al., 1991). We have extracted
150 an estimate of mean age for our North Atlantic transect from a recent inversion of ventilation tracer
151 observations (^{14}C , CFCs, PO_4^* , temperature and salinity) by Khatiwala et al. (2012). These
152 ventilation ages, which represent the time since a water parcel was last at the surface, taking into
153 account contributions from multiple pathways and source regions, are referred to in the text as
154 mean ages.

155 In addition to consideration of water mass ageing, we put our transect into hydrographic
156 context with the salinity and neutral density (γ_n) section in Fig. 1. The dome of salty subtropical
157 mode water, also known as Eighteen Degree Water, is apparent in the upper 500-800 m and is
158 roughly bound at depth by $\gamma_n = 26.65 \text{ kg m}^{-3}$ (LeBel et al., 2008). The remaining density surfaces
159 in Fig. 1 demarcate the boundaries between the various sources of NADW, which are defined most
160 clearly in the Northwest section between Bermuda and Woods Hole, Mass., known as Line W
161 (Toole et al., 2011). These are, in order of increasing density, Upper and Classic Labrador Sea
162 Water, Iceland-Scotland Overflow Water, and Denmark-Strait Overflow Water, which is underlain

163 by Antarctic Bottom Water (AABW, $\gamma_n > 28.125$). While we name the densest layer of water in
164 the western basin AABW, this water mass, far from its source, must have gone through significant
165 mixing with the overlaying NADW.

166 The deep waters of the Northeastern Atlantic (>3 km depth) are not as clearly defined by
167 the contributions to NADW and are characterized by a relatively homogeneous water mass called
168 Northeast Atlantic Deep Water (NEADW). NEADW is sourced by a mixture of NADW and
169 AABW which enters the Northeast basin largely through the Vema Fracture Zone at 11°N
170 (McCartney et al., 1991), with some contribution from the Romanche Trench near the equator
171 (Broecker et al., 1980; Schlitzer, 1987; Schlitzer et al., 1985). The intermediate water in the
172 southeastern portion of the cruise track intersects the northern extent of the salinity minimum (and
173 silicic acid maximum) originating from Antarctic Intermediate Water (AAIW) (Talley, 1999;
174 Tsuchiya, 1989), outlined in Fig. 1. Lastly, the high salinity intrusion of Mediterranean Outflow
175 Water (MOW) at ~1 km depth is well represented on the largely south-north part of the transect
176 approaching Portugal.

177 *1.3 Bottom scavenging*

178 Deep water ^{231}Pa and ^{230}Th concentrations can also be perturbed by changes in scavenging
179 intensity near the seafloor (bottom scavenging) associated with a change in particle concentration
180 or particle composition. Nepheloid layers (Biscaye and Eittrheim, 1977; McCave, 1986), or zones
181 up to hundreds of meters above the seafloor of increased particle concentration caused by the
182 resuspension of sediments, have been known to enhance the scavenging of the shorter-lived ^{234}Th
183 (half-life 24.1 days) in the northwest (Bacon and Rutgers v. d. Loeff, 1989; DeMaster et al., 1991)
184 and northeast (Schmidt, 2006; Turnewitsch et al., 2008; Turnewitsch and Springer, 2001) Atlantic.
185 Previous studies in the North Atlantic have suggested that bottom scavenging could reduce the

186 ^{230}Th concentration in deep water, but since the same effect can be achieved via recent water mass
187 ventilation without invoking a change in scavenging intensity, ventilation was the preferred
188 explanation (Moran et al., 1997; Moran et al., 1995; Vogler et al., 1998). However, recent results
189 from the Pacific, where the ventilation effect is not large enough to produce observed radionuclide
190 depletions in deepwater, have confirmed early observations (Bacon and Anderson, 1982; Nozaki
191 and Nakanishi, 1985) that significant bottom scavenging indeed occurs for ^{230}Th (Hayes et al.,
192 2013; Okubo et al., 2012; Singh et al., 2013) and ^{231}Pa (Hayes et al., 2013). Furthermore, nepheloid
193 layers in the South Atlantic have been found to significantly enhance scavenging of ^{230}Th and ^{231}Pa
194 (Deng et al., 2014).

195 Based on extensive observations in the northwest Atlantic of thick nepheloid layers
196 (Biscaye and Eittrheim, 1977; Brewer et al., 1976), our transect is well situated to determine the
197 effect of sediment resuspension on ^{230}Th and ^{231}Pa . In addition to increased particle loading,
198 bottom scavenging may also be affected by a change in particle composition. This section is also
199 well suited to test the hypotheses that ^{230}Th and ^{231}Pa are scavenged especially efficiently by
200 authigenic iron and manganese oxide phases associated with hydrothermal activity at the mid-
201 Atlantic ridge (German et al., 1991; German et al., 1993) or by (oxy)hydroxide coatings of particles
202 formed in regions of organic-rich sediment diagenesis at ocean margins (Anderson et al., 1983;
203 Bacon et al., 1976; Shimmield et al., 1986). To infer likely changes in scavenging intensity in our
204 transect, we utilize the distribution of the particle beam attenuation coefficient, C_p , as measured
205 by transmissometer from CTD casts, which is, to first order, linearly related to particle
206 concentration (Bishop, 1986; Gardner et al., 1985), although the sensitivity of C_p to particle
207 concentration is known to vary with particle size and composition (Baker and Lavelle, 1984;
208 Richardson, 1987).

209 **2. Methods**

210 The U.S. Geotraces North Atlantic transect (Fig. 1) consisted of two legs, collectively
211 designated GA03 in the global GEOTRACES survey (geotraces.org). KN199-4 (referred to as
212 GT10) from Lisbon, Portugal to Mindelo, Cape Verde was completed in Oct-Nov 2010. KN204-
213 1 (referred to as GT11) from Woods Hole, Massachusetts to Praia, Cape Verde via St. Georges,
214 Bermuda was completed in Nov-Dec 2011. Radionuclide data were produced by three
215 collaborating laboratories which were intercalibrated (Anderson et al., 2012) to analyze dissolved
216 (<0.45 μm) and particulate (0.45-51 μm) ^{232}Th , ^{230}Th , and ^{231}Pa in seawater: the Lamont-Doherty
217 Earth Observatory of Columbia University (L-DEO), the Woods Hole Oceanographic Institution
218 (WHOI) and the University of Minnesota (UMN). Five liter water samples were collected using
219 conventional Niskin bottles, filtered with 0.45 μm AcropakTM-500 filter capsules, and acidified to
220 pH = 1.8 at sea for storage according GEOTRACES protocols. Particulate samples representing
221 55-350 L of seawater were collected by McLane Research in situ pumps with a redesigned filter
222 holder (Lam and Morris, 2013) using paired 0.8 μm Pall Supor800 polyethersulfone filters
223 (Bishop et al., 2012).

224 Th and Pa isotopes (including the added tracers ^{229}Th and ^{233}Pa) were co-precipitated with
225 Fe (oxy)hydroxide for pre-concentration and purification using acid digestions ($\text{HNO}_3/\text{HF}/\text{HClO}_4$,
226 depending on the laboratory) and ion exchange chromatography. Filter samples were co-
227 precipitated with Fe after complete dissolution ($\text{HNO}_3/\text{HClO}_4/\text{HF}$). Radionuclide concentrations
228 were determined by isotope dilution inductively-coupled plasma mass spectrometry. We converted
229 radionuclide mass concentrations to radioactivity units using the conversion factors, 0.7591
230 $\mu\text{Bq}/\text{fg}$ ^{230}Th and 1.7476 $\mu\text{Bq}/\text{fg}$ ^{231}Pa . The analytical procedures used at L-DEO, WHOI, and

231 UMN have been fully described by Anderson et al. (2012), Auro et al. (2012), and Shen et al.
232 (Shen et al., 2003; Shen et al., 2002; Shen et al., 2012), respectively.

233 We correct measured dissolved radionuclide concentrations for in-growth due to uranium
234 decay during sample storage (Robinson et al., 2004). In-growth during sample storage from
235 particulate U concentrations is negligible (Anderson, 1982). In order to isolate the signature of
236 scavenging in the dissolved phase we also correct ^{230}Th and ^{231}Pa concentrations for a contribution
237 produced by the partial dissolution of U-containing lithogenic material based on dissolved ^{232}Th
238 as described by Hayes et al. (2013), assuming a crustal $^{238}\text{U}/^{232}\text{Th}$ ratio and congruent dissolution
239 of ^{232}Th , ^{230}Th and ^{231}Pa . Similarly, in the particulate phase we correct measured ^{230}Th and ^{231}Pa
240 for a lithogenic component based on particulate ^{232}Th . All radionuclide concentrations discussed
241 in the text are corrected for lithogenic sources and are denoted as “xs”. For more information on
242 data analysis see the metadata associated with these data online ([http://www.bco-](http://www.bco-dmo.org/dataset/3847)
243 [dmo.org/dataset/3847](http://www.bco-dmo.org/dataset/3847) or <http://www.bodc.ac.uk/geotraces/data/>). Forthcoming studies will
244 present and interpret the distribution of ^{232}Th and particulate radionuclides in their own right.
245 These results are used here only for the interpretation of the dissolved (or total) ^{230}Th and ^{231}Pa
246 distributions.

247 **3. Results and Discussion**

248 *3.1 Sections of dissolved ^{230}Th xs and ^{231}Pa xs*

249 Deviations from linear concentration-depth profiles as predicted by the model of reversible
250 scavenging are immediately apparent in the North Atlantic sections (Fig. 3). Both radionuclides
251 display substantial lateral concentration gradients, some of which are clearly related to recent
252 ventilation. Generally, lower concentrations of both radionuclides are found in the western and
253 northern parts of the transect, coincident with younger mean ages (Fig 3E). Additionally low

254 concentrations of ^{231}Pa xs in shallow water (Fig. 3D) take on a dome structure, coinciding with
255 EDW, and are presumably reflective of the rapid (<10 yrs) ventilation of this subtropical mode
256 water (Jenkins, 1988).

257 Notably unrelated to any change in mean age (Fig. 3E), ^{230}Th xs also has reduced
258 concentrations throughout the water column in the section between Cape Verde and Mauritania
259 (Cape Verde transect, Fig. 3B) coincident with increasing particle concentrations (Fig 3A, 5000-
260 6500 km section distance). The plunging isolines of ^{230}Th xs concentration toward the continental
261 margin on this transect are strong evidence for the process of boundary scavenging occurring. We
262 quantify the lateral transport of ^{230}Th xs and ^{231}Pa xs associated with this boundary scavenging in
263 section 3.2.

264 Both radionuclides show vertical concentration anomalies as well. Concentrations
265 generally increase linearly from the surface to depth but nearly always begin to decrease toward
266 the seafloor. These negative deviations with respect to reversible scavenging generally start higher
267 in the water column for dissolved ^{231}Pa xs (2-3 km depth) than for dissolved ^{230}Th xs (4-5 km).
268 This is a pervasive feature in the Atlantic (Luo et al., 2010; Moran et al., 2002; Scholten et al.,
269 2008; Scholten et al., 2001; Vogler et al., 1998) which has been largely attributed to the advection
270 of NADW in previous work.

271 At stations GT-10-01, GT11-04, GT11-06, GT11-08, and GT11-10, however, the dissolved
272 radionuclide depletions can be clearly associated with a large increase in beam attenuation related
273 to higher particle concentration (Fig. 3A) and presumably bottom scavenging. Additionally, the
274 near-bottom waters at GT11-16 (mid-Atlantic ridge) and GT10-09 (African margin) have a more
275 modest increase in C_p , but very large dissolved phase depletions. The near-bottom particles at these
276 two sites showed a clear enrichment in metal oxides (Lam et al., this issue), the former being up

277 to 40% authigenic Fe oxides from a hydrothermal plume at the mid-Atlantic ridge, the latter being
278 enrichment of authigenic Fe and Mn oxides (each 2-3% of the particle mass) related to reducing
279 conditions in the surface sediments created by organic matter diagenesis. It thus seems likely that
280 bottom scavenging due to increased particle abundance and/or unique particulate chemistry is at
281 least as significant as ventilation in regulating the distributions of dissolved ^{230}Th xs and ^{231}Pa xs
282 across the North Atlantic. In section 3.3, we give a few examples of how ventilation and bottom
283 scavenging may be convolved in determining ^{230}Th xs and ^{231}Pa xs distributions. The de-
284 convolution of these effects requires sensitivity testing in 3-dimensional ocean models which is
285 beyond the scope of this study.

286 *3.2 Quantification of boundary scavenging*

287 Enhanced removal of trace elements at ocean margins is supported by advective and
288 diffusive fluxes that arise due to the lateral concentration gradients imposed by lateral gradients in
289 scavenging intensity (Bacon, 1988). To quantify the magnitude of this flux, one can calculate the
290 lateral concentration gradients from concentration profiles. Since radionuclides are exchanged
291 between dissolved and adsorbed forms faster than they are removed to the seafloor (Bacon and
292 Anderson, 1982), for mass continuity, one must consider the total radionuclide concentration
293 (dissolved plus particulate). In Figure 4, we plot particulate and total ^{230}Th xs and ^{231}Pa xs for the
294 stations between Mauritania and GT11-22. Total ^{230}Th xs concentrations (Fig. 4C) are consistently
295 lower at stations closer to the African margin, at nearly all depths. The lateral gradient in total
296 ^{231}Pa xs (Fig. 4D), on the other hand, is smaller than can be resolved within our analytical
297 uncertainties. These observations are consistent with the boundary scavenging concept (Fig. 2). At
298 the margin, ^{230}Th concentrations can be depleted with respect to ocean interior concentrations to a
299 greater extent than ^{231}Pa concentrations. This is due to the longer ocean residence time of ^{231}Pa .

300 Lateral mixing and advection is not fast enough to erase the margin-interior ^{230}Th concentration
 301 gradient imposed by scavenging, whereas a lateral flux toward the margin is supported for ^{231}Pa .
 302 Consequently, both dissolved and particulate $^{231}\text{Pa}/^{230}\text{Th}$ xs ratios are higher at the margin versus
 303 the interior (Fig. 4E-F), as predicted by Bacon (1988).

304 In one dimension (x, an isopycnal surface, since circulation occurs preferentially along
 305 lines of constant seawater density), for total ^{230}Th xs (Th), the steady-state mass balance is:

$$306 \quad \frac{dTh}{dt} = P - S \frac{dTh_p}{dz} - u \frac{dTh}{dx} + K_H \frac{d^2Th}{dx^2} \quad \text{Eq. 1}$$

307 P is production due to ^{234}U decay. S is the particle sinking rate which, when multiplied by the
 308 vertical gradient of particulate ^{230}Th concentration (second term on right-hand side of Eq. 1),
 309 represents the downward flux by scavenging. The third and fourth terms on the right-hand side of
 310 Eq. 1 represents lateral fluxes due to advection (isopycnal velocity, u, multiplied by the first
 311 isopycnal concentration gradient) and eddy diffusion (isopycnal eddy diffusion coefficient, K_H ,
 312 multiplied by the second isopycnal concentration gradient), respectively.

313 In order to affect the steady-state mass balance, the advective and diffusive terms must
 314 occur on a timescale appropriate to the residence time, τ , of the tracer. The residence times, as
 315 defined by the water column inventory of radionuclide divided by its integrated production in the
 316 water column, between GT10-09 and GT11-22 for ^{230}Th and ^{231}Pa , respectively, are 10-28 yrs and
 317 120-150 yrs. The corresponding length scales over which lateral eddy diffusivity should be
 318 considered are ~800-1300 km for ^{230}Th and 2750-3080 km for ^{231}Pa ($\Delta x = \sqrt{2K_H\tau}$, parameterizing
 319 eddy diffusivity as a random walk process), using $K_H = 10^3 \text{ m}^2 \text{ s}^{-1}$, as determined in the Northeast
 320 Atlantic (Ledwell et al., 1998). The advective length scale ($\Delta x = u\tau$) depends linearly on the
 321 current speed and will be >1500 km for both radionuclides if u is greater than a typical deep current
 322 speed of 2 mm/s. However, because east-west velocities cyclically change direction in this region

323 (Zenk et al., 1991) and their magnitude is difficult to estimate for the deep ocean, we do not attempt
324 to quantify the advective flux.

325 The isopycnal gradients in ^{230}Th are estimated discretely using the observations from
326 GT10-09, GT10-10, GT10-11 and GT10-12 (Fig. 4C). We do not include GT11-22 in the analysis
327 because it makes the transect larger than the ^{230}Th -mixing length scale (~ 1400 km). The isopycnal
328 ^{231}Pa gradients are smaller than can be resolved within our analytical uncertainty. Th-230
329 concentrations were interpolated onto a common set of isopycnals (Fig. 5A), and because GT10-
330 11 and GT10-12 are nearly indistinguishable we average these two profiles and consider the
331 average profile representative of the region at the mid-point between the two stations.
332 Uncertainties were accounted for and propagated in the calculations by assuming a conservative
333 analytical uncertainty for total ^{230}Th xs of 1.5%. Two isopycnal gradients (Fig. 5B) were calculated
334 by differencing the concentration profiles, between GT10-09 and GT10-10 and between GT10-10
335 and GT10-11/12, and dividing by the lateral distance between the stations. A positive gradient is
336 defined as lower concentration in the east (leading to lateral fluxes toward Mauritania). Then the
337 second isopycnal gradient (Fig. 5C) was calculated by differencing the two isopycnal gradient
338 profiles and dividing by the distance between the mid-points of the stations used to calculate the
339 first gradient (Fig. 5D).

340 The second isopycnal gradient is variable above $\gamma_n = 27.8$ (1.2 km depth), but below this
341 density surface, in the bulk of the water column, the gradient is consistently positive (down
342 gradient toward Mauritania). By multiplying the $d^2\text{Th}/dx^2$ profile by K_H ($10^3 \text{ m}^2 \text{ s}^{-1}$), and
343 integrating with depth ($\sim 0\text{-}3$ km), we estimate the lateral convergence of ^{230}Th xs to the margin
344 (technically between two boxes, encompassing stations GT10-10/11/12 and GT10-09/10,
345 respectively) as $509 \pm 171 \text{ mBq m}^{-2} \text{ yr}^{-1}$. In 3 km of seawater, ^{230}Th production due to U decay is

346 1237 mBq m⁻² yr⁻¹ and thus the diffusive flux adds 41 ± 14% to the water column production in
347 the margin box (and removes the equivalent from production in the open-ocean box). This is
348 consistent with the upper limit for the model-derived redistribution of water column ²³⁰Th
349 (Henderson et al., 1999).

350 Th-230 normalized sediment fluxes will therefore be biased (underestimated) by up to 30-
351 50% for core sites at highly productive continental margins such as offshore Mauritania. The
352 magnitude of the complementary bias (overestimation) in ²³⁰Th-normalized fluxes in the interior
353 ocean is likely to be smaller than 30-50%. This is because the ²³⁰Th added to the relatively small
354 zone of high productivity at the margin is drawn from a much larger pool of the subtropical North
355 Atlantic gyre. The subtropical gyre ²³⁰Th budget, however, cannot be fully constrained here
356 because there are likely additional lateral removal fluxes of ²³⁰Th to other more expansive high
357 productivity regions such as the equatorial or subpolar North Atlantic (Henderson et al., 1999).

358 Interestingly, the degree of boundary scavenging (and its impact on lateral ²³⁰Th
359 redistribution) may have changed since the last glacial maximum (Lao et al., 1992). Nonetheless,
360 the 40% redistribution estimate can be seen as close to a global maximum (in the modern ocean at
361 least) since the Canary Current upwelling regime in which our observations are made produces
362 one of the largest lateral gradients in productivity (and in turn particle flux) in the world
363 (Behrenfeld and Falkowski, 1997). Thus our finding supports the use of ²³⁰Th-normalization to
364 reconstruct sediment fluxes within cited uncertainties (François et al., 2004).

365 Although we cannot directly estimate the magnitude of boundary scavenging for ²³¹Pa
366 because the lateral water column ²³¹Pa gradients are not discernible, we can use the ²³⁰Th results
367 in conjunction with the particulate ²³¹Pa/²³⁰Th xs data (Fig. 4F) to estimate the redistribution of
368 ²³¹Pa. The near-bottom ²³¹Pa/²³⁰Th xs ratio of particulate material, (Pa/Th)_{bottom}, should represent

369 the ratio of sinking flux for the two elements. If both elements were being buried at their production
370 rate by U decay, we would expect this ratio to be 0.093 (activity units, see dotted line, Fig. 4F).
371 Therefore the ratio of sinking flux to overlying production for ^{231}Pa , $(F/P)_{\text{Pa}}$, is proportional to that
372 of ^{230}Th , weighted by the deviation of the near-bottom particulate material from the production
373 ratio:

$$374 \quad (F/P)_{\text{Pa}} = (F/P)_{\text{Th}} * (\text{Pa/Th})_{\text{bottom}} / 0.093 \quad (\text{Eq. 2})$$

375 The average particulate $^{231}\text{Pa}/^{230}\text{Th}$ ratio of the two near bottom samples at GT10-09,
376 within about 100 m of the seafloor, is 0.18 (Fig. 4F). Using $(F/P)_{\text{Th}} = 1.4$ as calculated above, we
377 estimate that ^{231}Pa is being buried at 2.7 times its production in the overlying water column at
378 this site. While clearly a region of enhanced ^{231}Pa removal, this region will likely not account for
379 observed depletion of ^{231}Pa in deep North Atlantic sediment. High productivity areas such as
380 GT10-09, where boundary scavenging of ^{231}Pa is occurring, are likely only a small volumetric
381 percentage of the basin (Lippold et al., 2012b). This is indicated by the sharp decrease in particulate
382 $^{231}\text{Pa}/^{230}\text{Th}$ xs ratio between stations GT10-09 and GT10-10 (Fig. 4F). Nonetheless, more detailed
383 mapping of the extent of this type of enhanced ^{231}Pa burial on the North African margin is required
384 before its impact on the basin-wide Pa budget can be quantified (Burke et al., 2011; Marchal et al.,
385 2000).

386 *3.3 Bottom scavenging and ventilation: convoluted influences across the North Atlantic*

387 Both ^{231}Pa xs and ^{230}Th xs (Fig. 3) have negative concentration anomalies associated with
388 the high salinity waters of the Mediterranean Outflow Water (MOW, ~1 km depth at station GT10-
389 01, Fig. 1). This is expected due to the high particle load of MOW, a result of the interaction of
390 the flow of Mediterranean Water over the Gibraltar Strait and the Iberian margin (McCave and
391 Hall, 2002; Thorpe, 1972), which is known to enhance scavenging of ^{234}Th and ^{228}Th (half-life 1.8

392 yrs, parent ^{228}Ra) (Schmidt, 2006). Unexpectedly, the low radionuclide concentrations (causing
393 negative deviations to a linear profile) appear to be advected along with the flow of MOW to
394 station GT10-03 and even to GT10-05 for ^{231}Pa xs, while the high particle concentrations (on the
395 basis of C_p , Fig. 3A) are not, i.e. the downstream effects are not necessarily due to in-situ
396 scavenging. There is a strong boundary in ^{231}Pa xs concentrations and water mass age between
397 GT10-05 and GT10-07 at 2 km depth (Fig. 3), indicating more recently ventilated water to the
398 north, making it difficult to separate the downstream effects of specifically MOW from a large-
399 scale influence of other NADW components. This is the first of several examples of how
400 ventilation and bottom scavenging combine to produce low concentration anomalies in the water
401 column radionuclide distributions.

402 Second, we look in more detail (Fig. 6) at the impact of the enhanced scavenging observed
403 at station GT11-16, also known as the TAG hydrothermal site (Rona, 1980; Rona et al., 1984).
404 Clearly, dissolved ^{230}Th xs and ^{231}Pa xs are removed from solution in the observed hydrothermal
405 plume around 3.3 km depth (Figs. 3B, 3D, 6). But it also appears that the low concentration
406 anomaly in the plume is dispersed by circulation to shallower depths in the water column. The
407 TAG concentration profile departs negatively for dissolved ^{230}Th xs at 2.1 km, and for dissolved
408 ^{231}Pa xs at 1.5 km. Furthermore, the low radionuclide concentrations appear to be advected to sites
409 west of the ridge. For both radionuclides at 2.5 km depth, concentrations at GT11-14 are lower
410 than they are to the west at GT11-12, opposite to the expected trend due to water mass ageing.

411 Westward and shoaling propagation of the hydrothermal scavenging anomaly is consistent
412 with theory, i.e. buoyant plume water rising and heading west due to geostrophic considerations
413 (Speer, 1989). We note that this type of hydrothermal circulation will not be accounted for in the
414 mean age estimates. This type of “downstream” hydrothermal effects (both vertical and lateral,

415 Fig. 6) are not caused only by the observed venting at the TAG site but are more likely the
416 integrated result of vent sites all along the ridge (German et al., 2010). This result is support for
417 the hypothesis that hydrothermal vents are a basin-scale sink of Pa in the deep ocean (Hayes et al.,
418 2013). However, because the observed hydrothermal anomaly and its far-field effect occur in a
419 region with strong zonal and meridional gradients in water mass age, it is difficult to remove the
420 influence of ventilation and isolate the hydrothermal scavenging magnitude. For instance, one
421 could estimate the removal flux of ^{231}Pa (or ^{230}Th) by using the deficit of the observed
422 concentration profile compared to a linear profile (expected due to reversible scavenging) (Deng
423 et al., 2014). The expected profile however cannot be chosen *a priori*, because in a region of recent
424 deep water ventilation one does not expect a linear profile.

425 Similarly, along Line W (GT11-01 thru GT11-10), dissolved ^{230}Th xs and ^{231}Pa xs
426 concentrations are clearly depleted near the bottom (Fig. 3B, D) due to increased particle
427 concentrations in nepheloid layers (Fig. 3A). Depletion of ^{230}Th and ^{231}Pa below 3.5 km depth
428 cannot be attributed to ventilation, the conventional explanation (e.g., Luo et al., 2010), because
429 ventilation time scales increase with depth below 3.5 km (Fig. 3E). Nonetheless, the fact that ^{230}Th
430 and ^{231}Pa concentrations at mid-depth (1-3km) along Line W, the depth range of maximum
431 southward NADW transport (Cunningham et al., 2007; Kanzow et al., 2010; Talley et al., 2003),
432 are much lower than at the corresponding depths in the eastern basin at GT11-20 and GT11-22
433 (Fig. 3B, D), where particle concentrations (C_p values) are similar to those along Line W (Fig.
434 3A), seems clearly related to the east-west gradient in mean age. Radionuclide depletions due to
435 bottom scavenging and recent ventilation again here seem convolved in a way that is difficult to
436 untangle with static tracer observations.

437 In yet another combination of influences, dissolved ^{230}Th xs and ^{231}Pa xs concentrations
438 both decrease toward the bottom at GT11-20 and GT11-22 at ~4 km and 2.5 km depth,
439 respectively, where there is no evidence for increased particle concentrations or recent ventilation.
440 While the deep water in the Northeast Atlantic has been long isolated from the atmosphere (>500
441 yrs), it may not have been this long since a significant scavenging event occurred. The inflow of
442 NEADW from the Vema and Romanche Fracture Zones occurs on a timescale of 30 years based
443 on radiocarbon distributions (Schlitzer et al., 1985). The mid-Atlantic ridge is associated with
444 enhanced turbulent mixing because of its complex topography (Polzin et al., 1996) and with
445 metalliferous sediments from hydrothermal activity. Both of these factors could cause bottom
446 scavenging of ^{230}Th and ^{231}Pa from water flowing through the gaps in the ridge, the former via
447 resuspension of sediments and the latter by the increased scavenging efficiency of Fe-Mn oxides.
448 A radionuclide depleted signal could then be advected northward, carrying with it its scavenging
449 history. While this scenario is highly speculative, such are the possible interactions between deep
450 water flow and bottom scavenging that need to be accounted for in a fuller understanding of the
451 marine cycling of ^{230}Th and ^{231}Pa .

452 *3.4 Apparent controls on seawater and sedimentary $^{231}\text{Pa}/^{230}\text{Th}$ ratios*

453 Sensitivity tests in a scavenging-circulation ocean model, which can vary bottom
454 scavenging intensity and ventilation timescales independently, may be able to resolve the
455 significance of each process in determining radionuclide distributions in the North Atlantic. This
456 uncertainty notwithstanding, it is informative to inspect the section of the dissolved $^{231}\text{Pa}/^{230}\text{Th}$ xs
457 ratio (Fig. 3C) with a motivation to examine its proxy applications. High ratios along the Cape
458 Verde transect are consistent with boundary scavenging as discussed in section 3.2. In the areas of
459 clear bottom scavenging along Line W and at the TAG hydrothermal site, the ratio is also elevated

460 above the corresponding mid-depth values. This is because the ^{230}Th is scavenged more intensely
461 (larger depletion from the dissolved phase) relative to ^{231}Pa at these locations. In analogy to the
462 boundary scavenging due to lateral gradients in scavenging intensity, the bottom scavenging sites
463 may also act as preferential sinks for Pa. Because of its longer residence time, dispersive fluxes of
464 Pa into the bottom scavenging sites likely result in a greater Pa burial rate than would occur in the
465 absence of bottom scavenging (Deng et al., 2014).

466 The impact of ventilation on the dissolved $^{231}\text{Pa}/^{230}\text{Th}$ xs distribution (Fig. 3C), however,
467 is not obvious. For instance, between 2-3 km depth (excluding GT11-16) the $^{231}\text{Pa}/^{230}\text{Th}$ ratio
468 changes very little between 500 and 4200 km section distance (Fig. 3C), along which the mean
469 age has a strong lateral gradient between roughly 80 yrs in the west and 400 yrs in the east. This
470 range in age is the exact time period in which one expects the strongest return to a steady-state of
471 ^{231}Pa concentrations after ventilation (Gherardi et al., 2010), according to a 1-dimensional mixing-
472 scavenging model (Moran et al., 2001; Rutgers v. d. Loeff and Berger, 1993). The dissolved
473 $^{231}\text{Pa}/^{230}\text{Th}$ xs ratio of course increases further toward the African margin as ventilation age also
474 increases, but this is primarily due to the stronger removal of dissolved ^{230}Th xs (boundary
475 scavenging, sec. 3.2) at the eastern margin.

476 The observed zonal gradient in dissolved $^{231}\text{Pa}/^{230}\text{Th}$ xs (Fig. 3C) is not consistent with
477 removal of ^{231}Pa by southward flow between 2-3 km depth, which one expects to be strongest in
478 the west (Wunsch and Heimbach, 2006, 2013). That said, we cannot refute that the large-scale
479 deficit of ^{231}Pa in North Atlantic sediments (Lippold et al., 2012a; Yu et al., 1996) is consistent
480 with some amount of zonally-integrated southward ^{231}Pa transport from the North Atlantic to the
481 South Atlantic. This interpretation is supported by our particulate $^{231}\text{Pa}/^{230}\text{Th}$ xs data being mostly
482 below the production ratio (Fig. 4F) and by transport analysis of recent water column data from

483 the South Atlantic (Deng et al., 2014). Whether the $^{231}\text{Pa}/^{230}\text{Th}$ distribution (as opposed to ^{231}Pa
484 itself) is also responding to a zonally integrated southward flow of NADW remains to be
485 demonstrated in a 3-D dimensional circulation-scavenging model.

486 For instance, in the deep basins (4-6 km depth), likely not influenced by the African margin
487 processes, the dissolved $^{231}\text{Pa}/^{230}\text{Th}$ xs ratio is higher on the eastern side of the mid-Atlantic ridge,
488 as expected from the trend in mean age (Fig. 3D), perhaps reflecting the integrated removal of
489 ^{231}Pa (relative to ^{230}Th) by southward flow throughout the water column. Additionally, the ratio
490 decreases with depth (below 1 km and away from bottom scavenging sites), most significantly at
491 the deep central basin stations GT11-12 and GT11-20. The decrease with depth of the $^{231}\text{Pa}/^{230}\text{Th}$
492 ratio in the Atlantic has been suggested to reflect the southward export of ^{231}Pa by deep water
493 circulation (Lippold et al., 2011; Luo et al., 2010) based on 2-dimensional ocean models which
494 average out the east-west gradient in ventilation observed in our transect (Fig. 3E), also suggesting
495 a zonally-integrated effect of ventilation.

496 The influence of circulation may interestingly be more significant in the upper 1-1.2 km.
497 High $^{231}\text{Pa}/^{230}\text{Th}$ xs ratios near the surface are consistent with ^{230}Th being more intensely
498 scavenged out of the mixed layer, but a secondary subsurface maximum in dissolved $^{231}\text{Pa}/^{230}\text{Th}$
499 xs ratio around 1 km depth is observed at nearly every station. In the section between the mid-
500 ocean ridge and Mauritania, the secondary dissolved $^{231}\text{Pa}/^{230}\text{Th}$ xs maximum overlaps with AAIW
501 (cf. Fig. 1). The persistence of the $^{231}\text{Pa}/^{230}\text{Th}$ maximum in the more northern parts of the transect,
502 however, does not support an association with AAIW. An alternative scenario is that a high
503 $^{231}\text{Pa}/^{230}\text{Th}$ ratios throughout shallow water (0-1200 m), related to more intense scavenging of
504 ^{230}Th , is overprinted with the strong minimum around 500 m depth which could be related to the
505 low ^{231}Pa content and rapid ventilation of EDW. Yet another possible scenario is that the

506 subsurface dissolved $^{231}\text{Pa}/^{230}\text{Th}$ xs maximum is related to the preferential regeneration of
507 dissolved ^{231}Pa released during diatom dissolution (since biogenic opal is a strong scavenger of
508 ^{231}Pa (Chase et al., 2002)). This is not supported, however, by either the silicic acid distribution
509 (except within AAIW, Fig. 7B) or the particulate opal distribution (Lam et al., submitted). In any
510 case, the possibility of ^{231}Pa (and perhaps $^{231}\text{Pa}/^{230}\text{Th}$) tracing shallow or intermediate water
511 circulation is worth further attention since its longer-lived removal timescale and uniform
512 production offers a complement to the traditional transient tracers of shallow circulation (^3H - ^3He ,
513 CFC's).

514 As a way of summarizing these observations we cross-plot the dissolved $^{231}\text{Pa}/^{230}\text{Th}$ xs
515 data with another potential circulation tracer measured on the same water samples, silicic acid.
516 The silicic acid distribution along our transect (Fig. 7B) has a resemblance to the inverse estimates
517 of mean age (Fig. 3E). Silicic acid is added to deep water through the dissolution of diatom opal
518 exported from surface water and accumulates with deep water age (Broecker and Peng, 1982).
519 Furthermore, its distribution is sensitive to the overturning circulation since NADW has very low
520 (preformed) silicic acid and southern-sourced waters (AABW and AAIW) have much higher end-
521 member concentrations (Sarmiento et al., 2007). There is a wide scatter in the relationship
522 between dissolved $^{231}\text{Pa}/^{230}\text{Th}$ xs and silicic acid (Fig. 7A) and the overall negative trend reflects
523 that fact the $^{231}\text{Pa}/^{230}\text{Th}$ ratio generally decreases with depth while silicic acid (and mean age)
524 increase with depth. At some given depths (color scheme of points in Fig. 7A), however,
525 $^{231}\text{Pa}/^{230}\text{Th}$ may be positively related with silicic acid. For instance, around 1 km depth (dark blue
526 points in Fig. 7A) a positive trend possibly related to the influence of AAIW can be seen. Some of
527 the high $^{231}\text{Pa}/^{230}\text{Th}$ values at 3-4 km depth, however, are related to locations of bottom scavenging
528 (marked in Fig. 7A) and therefore cannot be ascribed to water mass ageing.

529 While a basin-scale, integrated influence of the southward export of NADW cannot be ruled
530 out, the distribution of dissolved $^{231}\text{Pa}/^{230}\text{Th}$ appears to be insensitive to water mass age across the
531 North Atlantic. In order to validate the use of the $^{231}\text{Pa}/^{230}\text{Th}$ ratio as a quantitative paleo-indicator
532 of the AMOC, a more complex conceptual model needs to be developed, including the influences
533 of boundary scavenging and bottom scavenging demonstrated here.

534 **4. Summary**

535 The cycling of ^{230}Th and ^{231}Pa in the ocean is complex. Deviations from the behavior expected
536 from a simple model of reversible scavenging are apparent across the North Atlantic and improved
537 spatial resolution allows us to study them in greater detail than has been done before. Boundary
538 scavenging of ^{230}Th in an exceptionally productive region off Northwest Africa can be constrained
539 to $40 \pm 10\%$ of its water column production, helping to quantify the uncertainties associated with
540 ^{230}Th -normalized sediment fluxes. Enhanced removal of ^{231}Pa occurs on the Africa margin as well
541 but quantitative conclusions about the significance of this sink in the basin-scale Pa budget cannot
542 be made without more detailed mapping of the region. Both recent ventilation and bottom
543 scavenging cause deep-water depletions of ^{230}Th and ^{231}Pa . The dissolved $^{231}\text{Pa}/^{230}\text{Th}$ ratio traces
544 locations of intense scavenging intensity while its distribution in the transect is not consistent with
545 a simple relationship to water-mass age. We observe several examples where the effects of
546 scavenging and ventilation are convolved which provide excellent test cases for sensitivity studies
547 of removal mechanisms in future ocean modeling of these isotopes. Circulation and scavenging
548 affect many trace metals of biogeochemical and paleoceanographic interest (e.g. Fe, Co, Al), and
549 thus further constraining the cycling of ^{230}Th and ^{231}Pa will be of broad appeal in the oceanographic
550 community.

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565

566 **Figure Captions**

567 **Figure 1** Map of GEOTRACES section GA03, the U.S. GEOTRACES North Atlantic Transect
568 and section of salinity as measured in the Niskin bottle rosette casts. Neutral density overlays
569 define, in order of increasing density, the bottom of Eighteen Degree Water (EDW), Upper and
570 Classic Labrador Sea Water (ULSW/CLSW), Iceland-Scotland Overflow Water (ISOW),
571 Denmark-Strait Overflow Water (DSOW) and Antarctic Bottom Water (AABW), defined largely
572 for the western basin by Toole et al. (2010) and LeBel et al. (2008). The deep (>3 km) eastern
573 basin is filled with a more homogeneous water mass named Northeast Atlantic Deep Water
574 (NEADW) (Schlitzer et al., 1985). The southeastern section is influenced by Antarctic
575 Intermediate Water (AAIW) at about ~1 km depth and the northeastern margin is clearly
576 influenced by the high salinity Mediterranean Outflow Water (MOW). Presented in this paper is
577 dissolved radionuclide data for all stations listed here, and a sub-set of particulate radionuclide
578 data for stations marked with a black diamond.
579

580 **Figure 2.** Schematic representation of boundary scavenging. Along distance from an ocean
581 margin, particle flux (in arbitrary units, a.u.) increases nearly exponentially. The particulate and
582 dissolved $^{231}\text{Pa}/^{230}\text{Th}$ ratio are expected to increase with increasing particle flux, associated with
583 increasing scavenging intensity. Note that the particulate $^{231}\text{Pa}/^{230}\text{Th}$ ratios overlap the
584 production activity ratio (A.R., 0.093) produced from uranium decay, whereas the dissolved
585 ratios are roughly 10 times higher due to the preferential scavenging of ^{230}Th from seawater. The
586 longer residence time of ^{231}Pa , compared to that of ^{230}Th , allows a greater lateral flux of ^{231}Pa
587 down the concentration gradient toward the margin. This results in increased sinking vertical flux
588 of ^{231}Pa at the margin, at the expense of a lower vertical flux in the ocean interior. In contrast, the
589 sinking ^{230}Th flux is relatively uniform since its lateral movement is more limited. Therefore the
590 water column sustains greater lateral concentration gradients in ^{230}Th than in ^{231}Pa . The
591 numerical values used here are simply for illustration and do not represent actual observations.
592

593 **Figure 3** Property sections of the US GEOTRACES North Atlantic transect. (A) Particle beam
594 attenuation coefficient, C_p , determined by transmissometer. CTD casts are marked in black. (B)
595 Dissolved (<0.45 μm) ^{230}Th , (C) $^{231}\text{Pa}/^{230}\text{Th}$ activity ratio, and (D) ^{231}Pa . Black dots indicate

596 discrete measurements. (E) Mean age of seawater since being at the surface as estimated by
597 Khatiwala et al. (2012). Station locations for radionuclide data are as labeled in the map of
598 Figure 1. Panels A and E, in addition, have data from shallow “demi” stations occupied in
599 between full depth stations, as plotted in the inset map of panel E. See Fig. 1 for neutral density
600 surfaces not included here for clarity.

601
602 **Figure 4** Depth profiles of particulate (0.45-51 μm) (A, B) and total (C, D) ^{230}Th xs and ^{231}Pa xs
603 and the dissolved ($<0.45 \mu\text{m}$) (E) and particulate (F) $^{231}\text{Pa}/^{230}\text{Th}$ xs ratio from stations along a
604 zonal transect between Mauritania and west of Cape Verde. The dashed lines in (E) and (F)
605 represent the activity ratio of $^{231}\text{Pa}/^{230}\text{Th}$ produced by uranium decay (0.093). Legend relates
606 station names as located in Figure 1.

607
608 **Figure 5** (A) Depth profiles of total ^{230}Th interpolated onto a common set of isopycnal surfaces.
609 (B) Discretely calculated lateral concentration gradients in total ^{230}Th . Units are $\mu\text{Bq} \times 10^{-3}$ per
610 kilogram seawater per kilometer distance (C) Discretely calculated second lateral concentration
611 gradient in total ^{230}Th . Units are $\mu\text{Bq} \times 10^{-6}$ per kilogram seawater per square kilometer (D)
612 Schematic demonstrating the concept of calculating lateral gradients using the concentration
613 difference between depth profiles and the distance separating the profile locations along the Cape
614 Verde transect.

615
616 **Figure 6** Depth profiles of dissolved ^{230}Th and ^{231}Pa from the hydrothermal TAG site, GT11-16,
617 and the surrounding stations. As inferred from the section (Fig. 3), it is possible that the strong
618 removal of both isotopes at hydrothermal plumes along the Mid-Atlantic Ridge causes downstream
619 radionuclide depletion, with respect to the linear increase in concentration with depth expected
620 from reversible scavenging, at sites to the west of the ridge (GT11-14 and GT11-12) but not
621 significantly to the east (GT11-18).

622
623 **Figure 7** (A) Dissolved $^{231}\text{Pa}/^{230}\text{Th}$ xs ratios versus silicic acid measured in the same samples. The
624 arrow indicates a possible positive trend between $^{231}\text{Pa}/^{230}\text{Th}$ xs ratios and silicic acid carried by
625 Antarctic Intermediate Water (AAIW). The circled points represent locations of bottom
626 scavenging which causes elevated $^{231}\text{Pa}/^{230}\text{Th}$ xs ratios and are unrelated to the circulation patterns
627 traced by silicic acid. (B) Silicic acid distribution along the North Atlantic transect.

628

629

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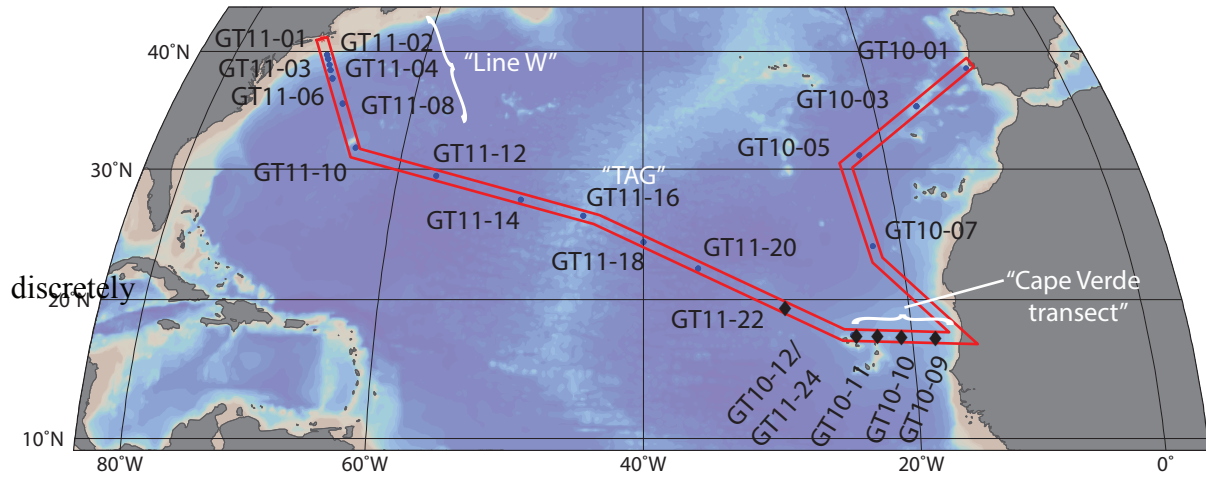
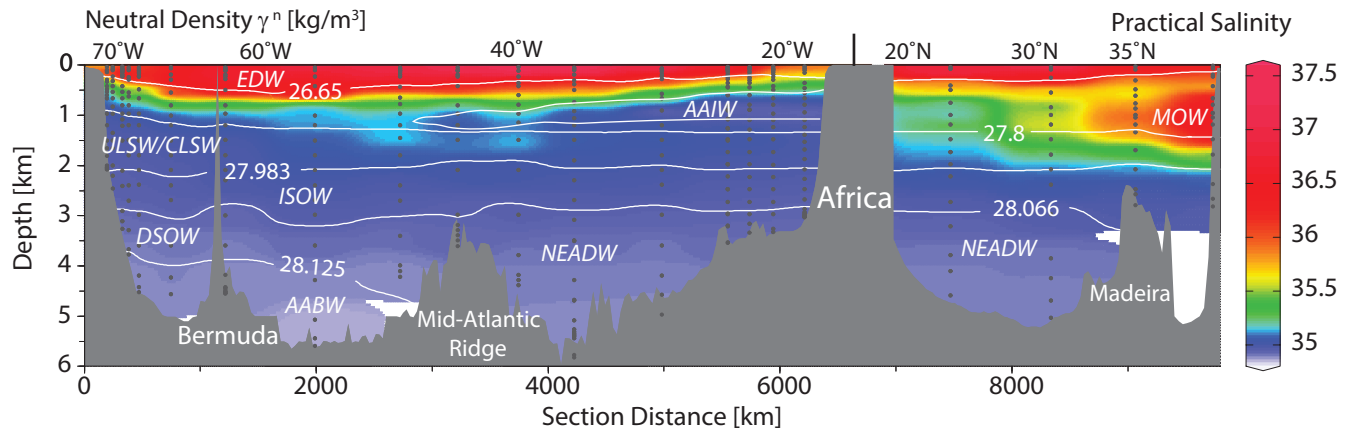
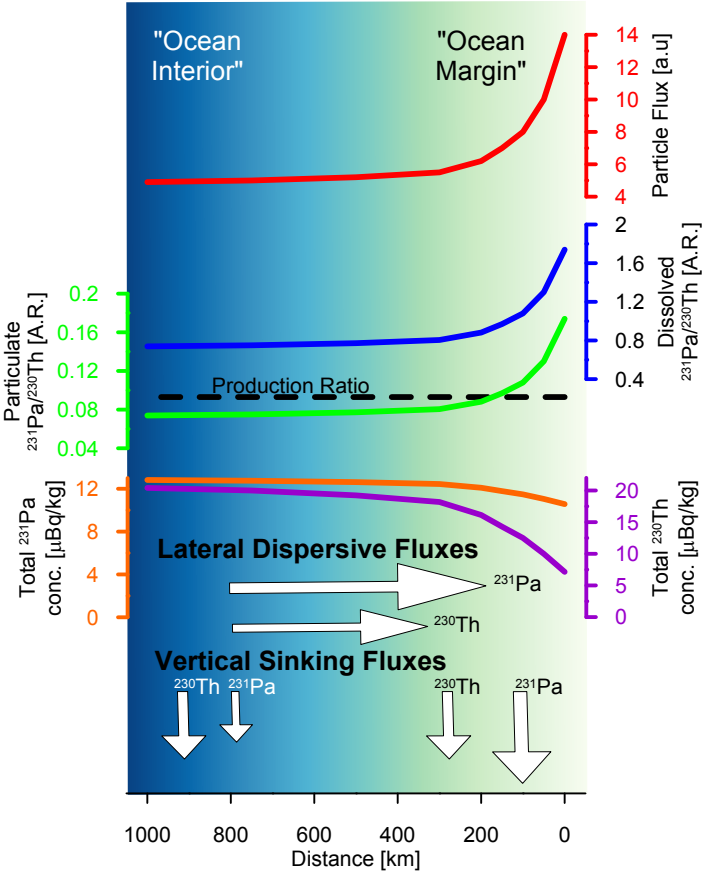


Figure 1



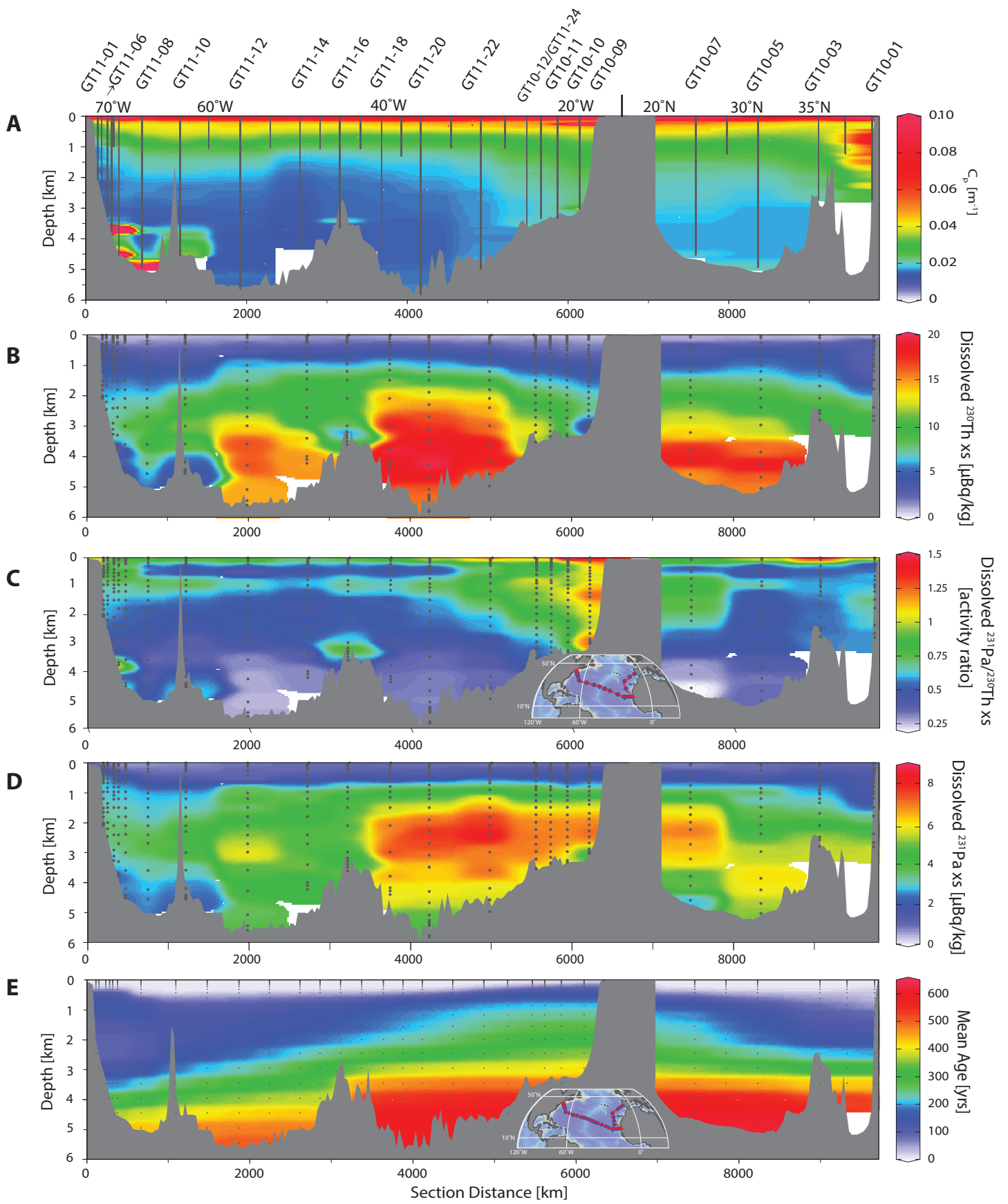
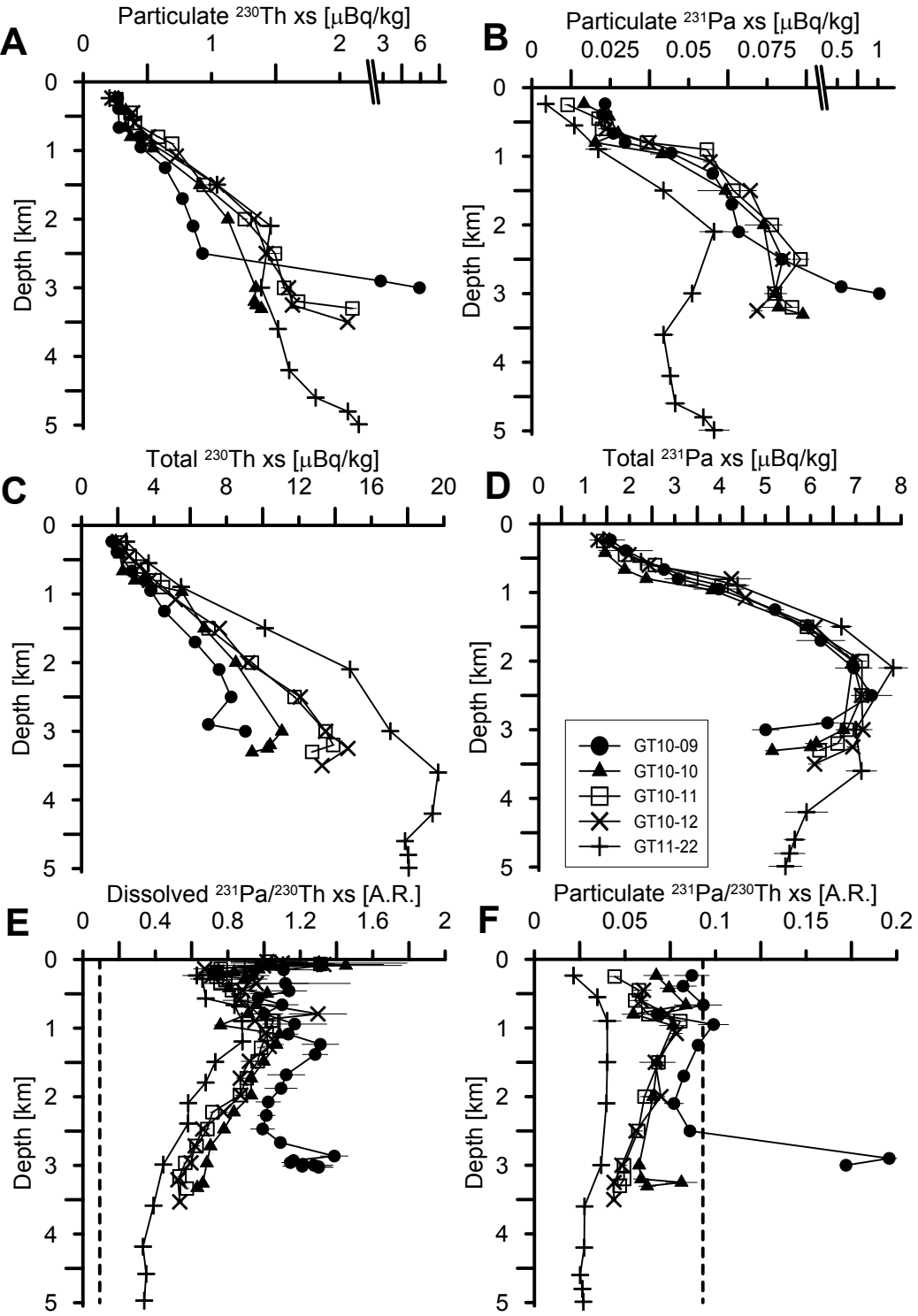
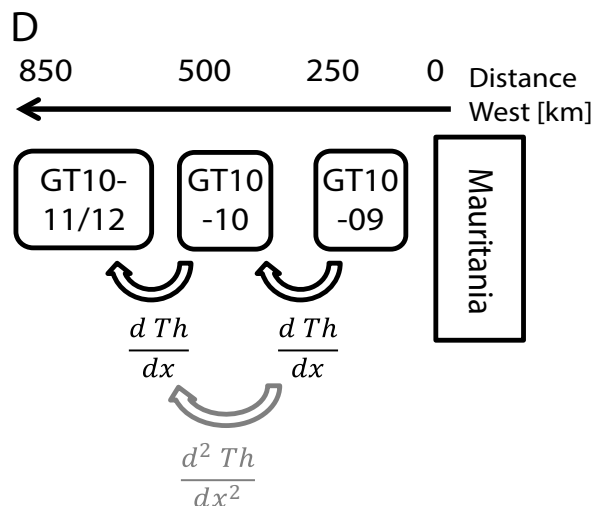
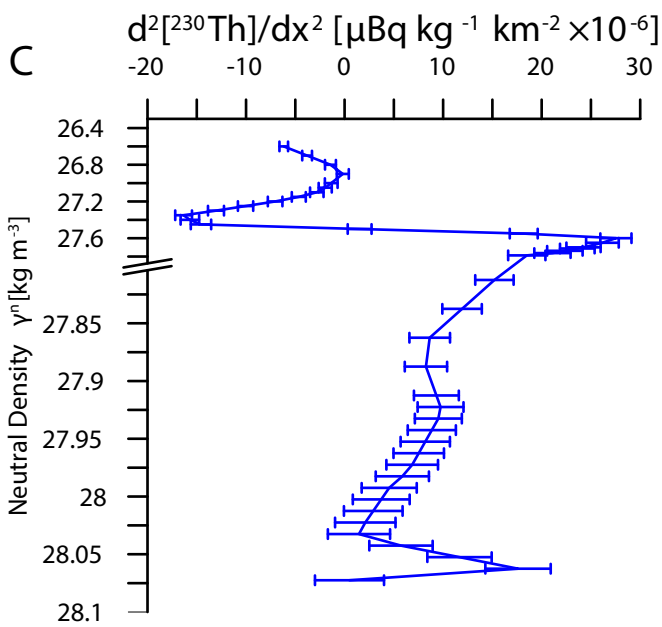
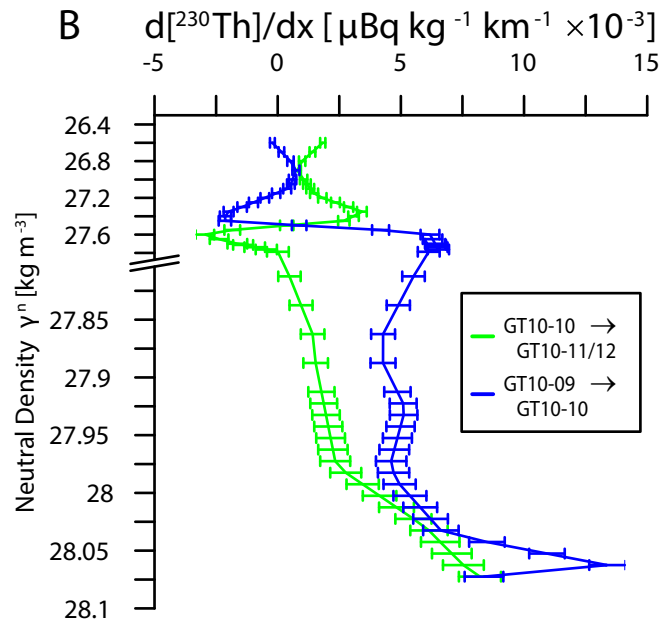
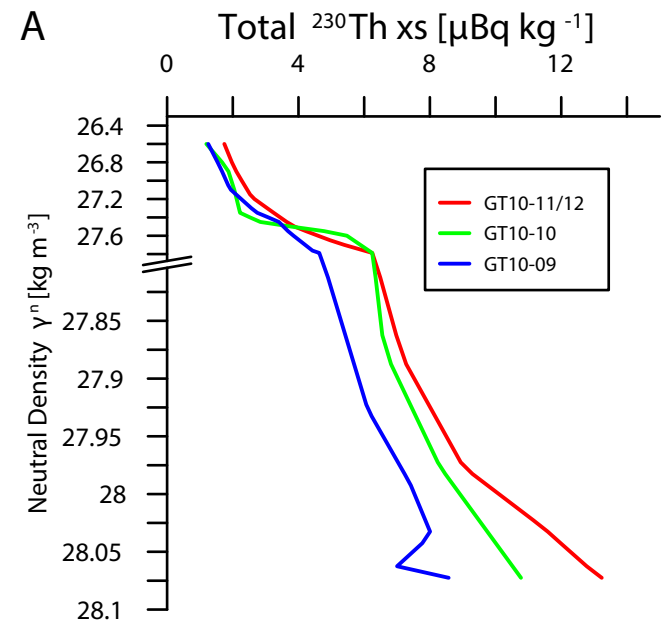
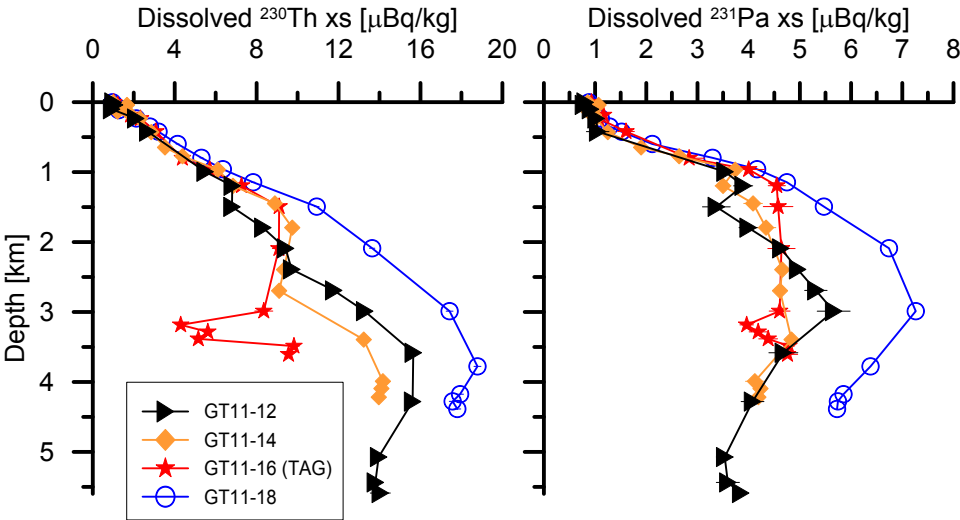


Figure 3.







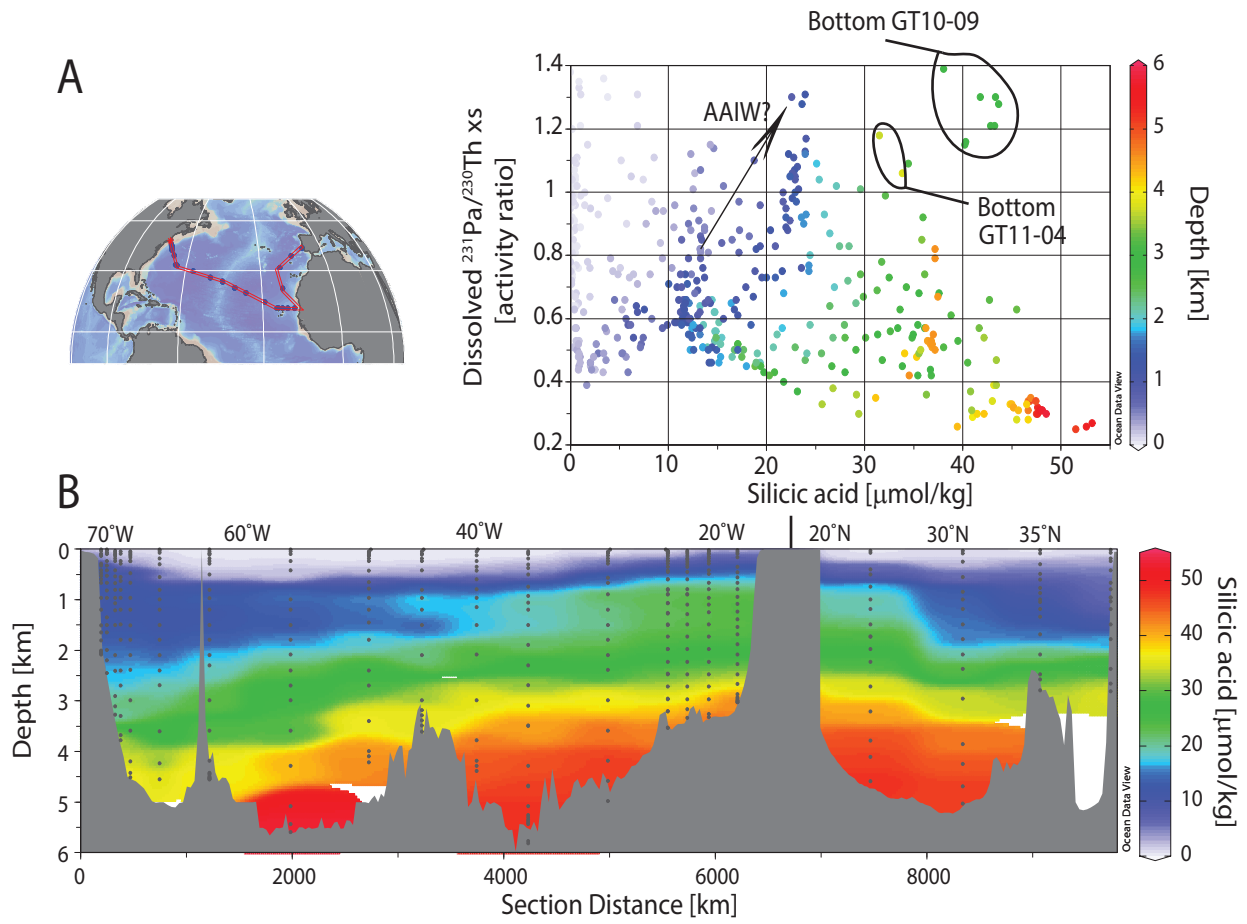


Figure 7.