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1 **Thorium isotopes in the Southeast Atlantic Ocean: Tracking scavenging**
2 **during water mass mixing along neutral density surfaces**

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31 **Abstract**

32 The distributions of dissolved and particulate thorium isotopes (^{230}Th and ^{232}Th) were
33 established in samples from the BONUS GoodHope (BGH) IPY-GEOTRACES cruise in the
34 SE Atlantic sector of the Southern Ocean (36°S - 13°E to 57°S - 0° , Feb.–Mar. 2008). The
35 distribution of total (dissolved+particulate) ^{232}Th is dominated by the inputs from continental
36 margins. The non-linear profiles of dissolved ^{230}Th are interpreted as due to the southward
37 upwelling of the isopycnal surfaces. However, total ^{230}Th and ^{232}Th versus salinity plots
38 illustrate departures from binary mixing and provides evidence for non-conservative behavior
39 of both isotopes along the section. We propose a model for total ^{230}Th and ^{232}Th scavenging
40 and mixing along isopycnal surfaces. We use this model to estimate particle settling speeds
41 and isopycnal eddy diffusion coefficients along the BGH section. Data-model comparison
42 suggests particle settling velocities in the range of 400-700 m/y and isopycnal eddy diffusivity
43 of the order of 2000 m^2/s .

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52 Keywords: thorium isotopes, Southern Ocean, Atlantic, marine particles, isopycnal mixing

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54 Highlights:

55 Small deviations of thorium isotopes from a conservative behavior occurs during isopycnal
56 mixing through the ACC.

57 An advection-diffusion-scavenging model along neutral density surface is applied to thorium
58 isotopes.

59 Combining thorium isotopes allows constraining particle settling speeds and isopycnal mixing
60 coefficients

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71 1. Introduction

72 Thorium isotopes are recognized tracers of particle dynamics in the ocean that provide
73 key information on ocean biogeochemical cycles (Bacon and Anderson, 1982, Nozaki et al.,
74 1981, Roy-Barman et al., 1996). In particular, ^{230}Th is produced *in situ* by radioactive decay
75 of ^{234}U and rapidly scavenged on marine particles, thus providing a chronometer for
76 estimating the settling speed for marine particles. ^{232}Th is introduced in the ocean by
77 continental inputs and hence can bring complementary information on marine particles
78 compared to ^{230}Th (Roy-Barman et al., 2002).

79 In early studies, particle settling speeds were derived from ^{230}Th with a simple 1D
80 (vertical) production-scavenging model, which was well adapted to oceanic areas with weak
81 currents (Bacon and Anderson, 1982, Nozaki et al., 1981, Roy-Barman et al., 1996, Roy-
82 Barman, 2009). However, it is now clear that strong currents characterizing oceanic regions
83 such as the Southern Ocean may significantly affect the distribution of ^{230}Th in the water
84 column and hence potentially bias the estimate of the settling speed of marine particles
85 obtained by the simple production-scavenging model. Therefore, a simple production-
86 scavenging-advection model was proposed to constrain simultaneously the renewal rate of the
87 deep waters and the settling speed of marine particles in the Atlantic sector of the Southern
88 Ocean (Rutgers van der Loeff et al., 1996). However, it relies on rather crude approximations,
89 such as uniform values for ventilation rate and particle settling speed over the whole water
90 column. Another limitation of this model is that it does not consider the mixing of water
91 masses with different origins and Th contents.

92 In the real ocean, the distribution of geochemical tracers depends on both non-
93 conservative processes and mixing of water masses of different origins, in addition to
94 advection by currents. This mixing is thought to occur preferentially along surfaces of
95 constant density called isopycnal surfaces. Chase et al. (2003) suggested that the small
96 gradients in ^{230}Th concentration observed along neutral density surfaces in the deep waters of
97 the Southern Ocean argue for small eddy diffusive fluxes and little scavenging of ^{230}Th across
98 the Antarctic Polar Front (APF). However, they also advocated for further work to better
99 constrain both nuclide concentrations along isopycnal surfaces and diffusion coefficients in
100 the Southern Ocean.

101

102 In the present work, we take advantage of high precision ^{230}Th data collected during
103 the Bonus GoodHope cruise to revisit the behavior of ^{230}Th across the Southern Ocean and to
104 try to unravel the respective roles of water mass mixing and particle scavenging along neutral

105 density surfaces. Our goal is to explore if Th data can help constraining both particle settling
106 speeds and water mixing rates using a model that is more complete than with the simple
107 mixing-scavenging model.

108

109

110 2. Sampling and methods

111 2.1. Hydrographic settings

112 The oceanographic structure determined from the Bonus GoodHope (BGH) expedition
113 has already been described (Chever et al., 2010, Bown et al., 2011, Garcia-Solsona et al., 2014,
114 Abadie et al., 2017), so we only summarize the main features and focus on the 5 super stations
115 (S1–S5 stations, Fig. 1) sampled for Th isotopes. Three main hydrological and circulation
116 regimes characterize the studied area: the Subtropical domain (Station S1), the Antarctic
117 Circumpolar Current (ACC; S2 to S4) and the Weddell Sea Gyre (S5). The subtropical domain
118 is bounded to the south by the subtropical front (STF), which was located around 41°S–42°S,
119 north of S2. The ACC domain, incorporating the Subantarctic Front (SAF; ~45°S) and the Polar
120 Front (PF; ~50°S), is bounded to the south by the southern ACC boundary (Sby). From the Sby
121 to the Antarctic continent, water circulation is dominated by the cyclonic Weddell Gyre (WG;
122 S5).

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127 The subtropical regime is influenced from 0 down to ~800 m by surface and
128 intermediate waters of Indian Ocean origin ($\theta \geq 10$ °C, $S \geq 34.5$), conveyed south of Africa by
129 the Agulhas Current and detached eddies (Gladyshev et al., 2008). At intermediate depths
130 (~300–900 m), Antarctic Intermediate Water (AAIW) originates from cooled surface waters of
131 the ACC that are subsequently subducted northward at the PF. Two varieties of AAIW have
132 been recognized from BGH cruise data: one originating from the Indian Ocean (i-AAIW) close
133 to the African continent ($S > 34.3$; S1) and one formed in the Atlantic Ocean (a-AAIW; $S <$
134 34.3 ; S2–S4). The Upper Circumpolar Deep Water (UCDW; ranging from 500 to 2000 m depth)
135 is identified below the AAIW and upwells southwards. Below UCDW, the eastward flowing
136 (and southward drifting) NADW is detected by its salinity maximum within the 2000–3000 m
137 depth range. Two NADW branches are identified in the BONUS section: North of the Agulhas
138 Ridge, higher salinities indicate advection by an eastern boundary current referred to as SE-

139 NADW (S1 and S2 stations). South of the Agulhas Ridge, the main (western) component of
140 NADW (i.e., SW- NADW; S2-S4 stations) is incorporated into the ACC after flowing
141 southward along the Argentine-Brazilian margin (Stramma and England, 1999). The other
142 water mass encountered at southern ACC latitudes is high salinity Lower Circumpolar Deep
143 Water (LCDW). Cold and fresher Antarctic Bottom Water (AABW) is also recognized,
144 occupying the water column from LCDW/NADW depths down to the sea floor (Fig. 2). In the
145 Weddell Sea Gyre domain (S5), we interpret deep oxygenated waters as due to input of recently
146 ventilated AABW.

147 Water mass transport along the BGH section is mainly zonal. In the subtropical domain
148 (station S1), it is dominated by the westward flowing Agulhas Current in the upper water
149 column ($z < 1500$ m) and at depth by the eastward transport of SE-NADW as delineated by
150 Sokolov and Rintoul (2009). Stations S2, S3 and S4 are located within the Antarctic
151 Circumpolar Current, flowing eastward throughout the water column. Finally station S5 is
152 located in the Northern limb of the Weddell gyre, also flowing eastward. Therefore, the BGH
153 section is approximately perpendicular to the climatological mean (dominant) flows, so that
154 water masses should not flow directly from one BGH station to another. The different BGH
155 stations would therefore not be directly connected by the mean flow. However, superimposed
156 on this vigorous zonal circulation, slower meridional transports are clearly visible on meridional
157 sections of water mass tracers such as salinity, temperature or nutrients, revealing notably the
158 northward transport of AAIW and AABW, and southward transport of NADW (Abadie et al.,
159 2017). This meridional transport is presumably a residual circulation in which lateral eddy
160 fluxes largely balance the wind-driven circulation (Marshall and Speer, 2016). While the
161 circulation in the BGH area is therefore clearly 3D and dominated by zonal flows (preventing
162 a direct link by mean advection between our 5 stations), some of the BGH samples can still
163 reflect the effect of these meridional transports of these water masses upstream of the BGH
164 section. In the following, transport models will be used to link tracer properties measured along
165 the BGH section. We do not assume that the transports actually act along the BGH section (pure
166 meridional transport is not realistic). However, we use the tracer properties measured along the
167 BGH section to elucidate and quantify the meridional component (advection, diffusion)
168 embedded within a dominant zonal component and therefore acts upstream of the BGH section,
169 for instance between the Drake Passage and the BGH section.

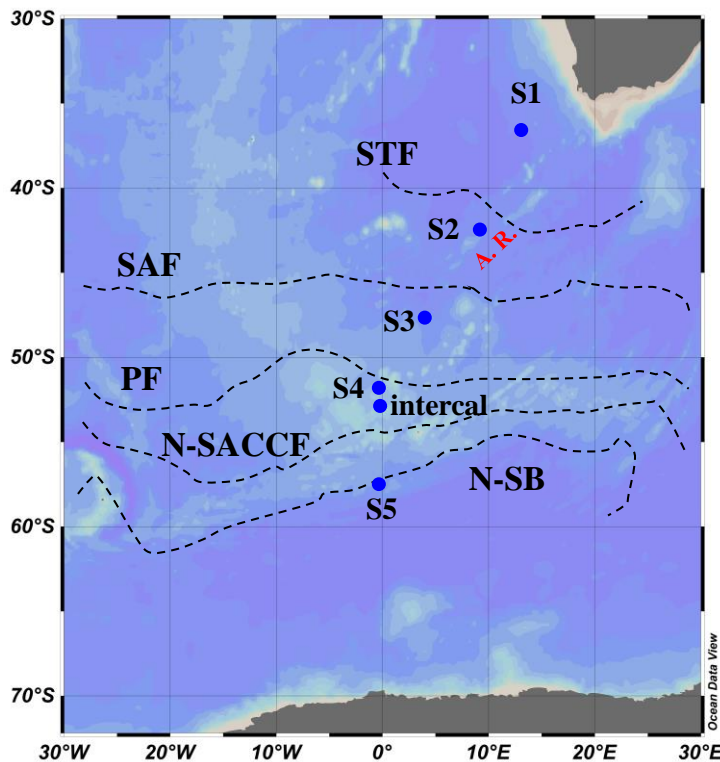
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172 2.2. Sampling

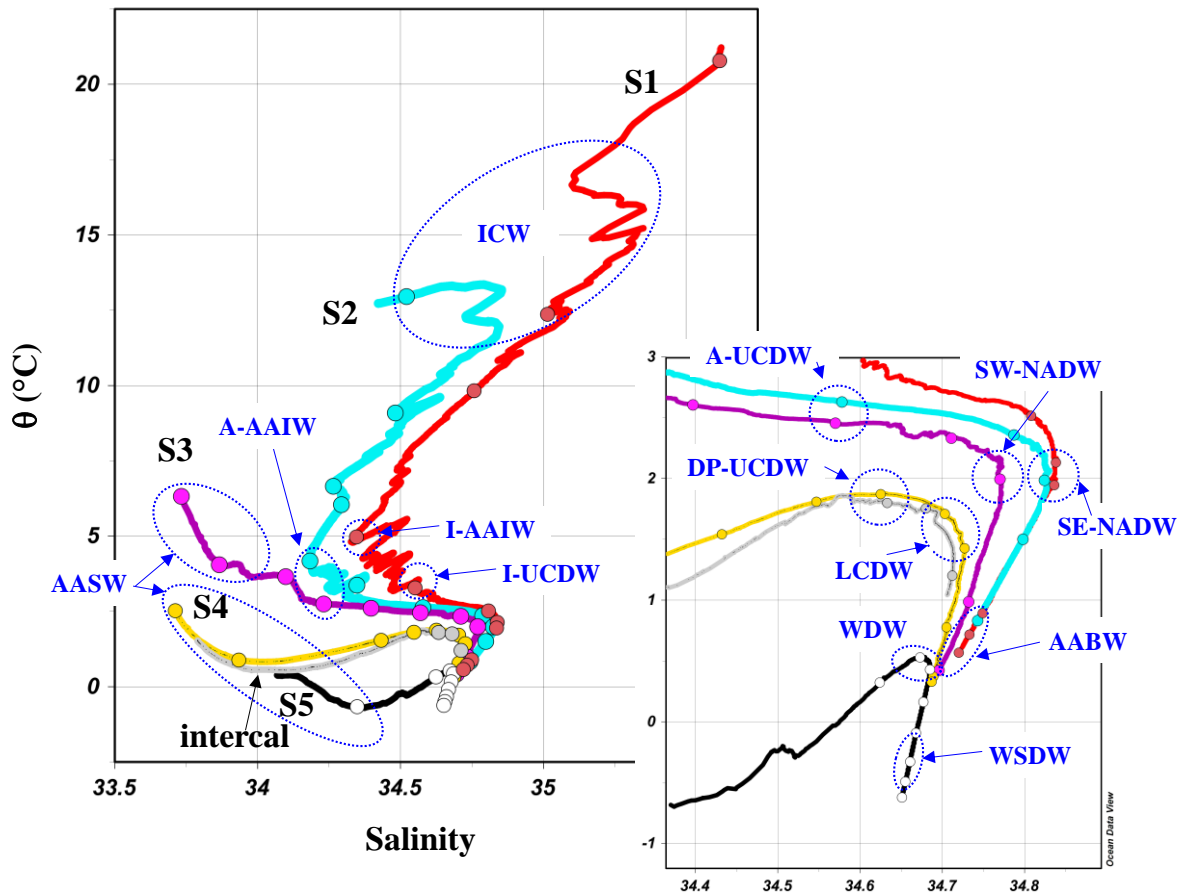
173 The BONUS GoodHope cruise took place in the framework of the international polar
 174 year (IPY) from February 8th 2008 till March 17th 2008 on board the Research Vessel Marion-
 175 Dufresne II, from 34°S 18°E to 51°S 00°W and from 51°S to 58°S along the Greenwich
 176 meridian. The expedition took place during the late austral summer, when biological activity is
 177 high and ice cover is reduced. Five depth profiles (S1–S5 stations) were sampled for both
 178 dissolved and particulate Th with 10 or 11 depths per profile with 1 duplicated sample at stations
 179 S3, S4 and S5 (Fig. 1). An intercalibration (hereafter referred to as ‘intercal’) station was
 180 analyzed for 3 depths which were all duplicated.

181 Seawater samples were collected using Niskin bottles equipped with Teflon coated
 182 strings and mounted on a rosette frame with a CTD sensor. Challenger Oceanic *in situ* pumps
 183 (ISP) were used to filter large seawater volumes through SUPOR filters (pore size: 0.45 µm,
 184 filter diameter: 293 mm). Typically, volumes ranging from 200 L to 1000 L were filtered.



185
 186 Fig. 1. Sampling stations during the BONUS-GoodHope section and main fronts (following
 187 Sokolov and Rintoul, 2009; mean path position): STF = SubTropical Front; SAF = SubAntarctic Front
 188 (middle branch); PF = Polar Front (middle branch); N-SACCF = northern branch of the Southern ACC
 189 front; N-SB = northern branch of the Southern Boundary. A.R.: Aghulas Ridge.

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193 Fig.2: Potential temperature-salinity data during BGH. (a): full depth profiles. (b): deep
 194 waters. Dots: data at the depth of dissolved Th samples. Acronyms: AABW, Antarctic Bottom
 195 Water; AASW, Antarctic Surface Water; I-AAIW and A-AAIW, Antarctic Intermediate
 196 Water of Indian and Atlantic origins; I-CW, Indian Central Water; I-UCDW, A-UCDW, and
 197 DPUCDW, Indian, Atlantic, and Drake Passage Upper Circumpolar Deep Water; LCDW,
 198 Lower Circumpolar Deep Water; SE-NADW and SW-NADW, Southeast and Southwest
 199 North Atlantic Deep Water; WDW, Warm Deep Water; WSDW, Weddell Sea Bottom Water;
 200 WW, Winter Water.

201

202 2.3. Analytical procedure

203 *Filtered seawater*

204 Twenty liters of filtered seawater (Nuclepore, 90 mm diameter, 0.4 μm pore size) were
 205 acidified, spiked with ²²⁹Th and pre-concentrated on board using the ferric hydroxide co-
 206 precipitation technique as described in Piepgras et al. (1979). The precipitate was then
 207 recovered on filters (Supor, 90 mm, 0.4 μm), which were kept in petri dishes and stored in the
 208 dark. Back in the laboratory, the filters were submerged in 40 mL of 6 M HCl for 24 h in 300
 209 mL Polytetrafluoroethylene beakers, enabling the dissolution of precipitate. The filters were
 210 then carefully rinsed with deionized water to recover all the dissolved precipitate. After

211 evaporation, the residue was dissolved in 0.5 mL 7M HNO₃ and loaded on an anion-exchange
212 column (AG1-X8 resin, 200–400 mesh) to separate the thorium fraction from the major
213 elements, uranium and the rare earth elements (REE) (adapted from Jeandel et al., 2011). The
214 total ²³²Th chemical blank is typically 48 ± 14 pg and represents generally less than 15% of the
215 sample although peaks with 50% of the sample occur for very ²³²Th-poor samples. Total ²³⁰Th
216 chemical blank is typically 2.3 ± 0.1 fg and represents generally less than 5 % of the sample
217 although peaks with 30% of the sample occur for very shallow samples.

218

219 *Particles collected from Niskin bottle filtrations*

220 From the twenty liter seawater samples used for dissolved Th analyses (described
221 above), particulate samples were also available on the 90mm diameter 0.4µm porosity
222 Nuclepore membranes used for filtration (hereafter referred to as Niskin filtration). Membranes
223 were transferred from their petri dishes to Teflon beaker containing 15mL 6M HCl and 2.5mL
224 15M HNO₃. They were heated 90 minutes at 130°C. After leaving them cool, 0.5mL 23M HF
225 was added, and the mixture was heated again as previously. The membranes were removed
226 from the leaching solutions and rinsed over the beakers with milliQ water. After evaporation,
227 the samples were dissolved in 3mL 0.32M HNO₃ for analysis by Quadrupole Induced Coupled
228 Plasma Mass Spectrometry.

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232 *Particles collected from in situ pump filtrations*

233 Filters from the in situ pumps were cut into pieces using ceramic scissors. Several
234 leaching steps were necessary to recover Th without attacking the filters. Filter pieces were first
235 leached with 200 mL of 6N HCl and 1.5 mL of 24-25N HF in a 300 mL Teflon beaker for 2
236 days at 75°C. The filter pieces were removed from the leaching solution and rinsed with a MQ
237 water squeeze bottle over the leaching beaker. The filter pieces were saved apart. The leaching
238 and rinsing solutions were evaporated down to ~10 mL and transferred into a 30 mL Teflon
239 beaker. Then, the filter pieces were leached a second time with 150 mL of 7N HNO₃ and 0.05
240 mL of 29N HF for 2 days at 75°C. The filter pieces were removed from the leaching solution
241 and rinsed with MQ water that was again recovered into the leaching beaker. After evaporation
242 to a few mL, this solution was added to the first leaching/rinsing solution. The 300 mL Teflon
243 beaker was rinsed in warm diluted HNO₃ to remove any particle sticking on the beaker walls
244 and the resulting solution was also added to the 30 mL beaker. The resulting solution was then

245 spiked with ^{229}Th (and ^{233}Pa), dried and dissolved again in a solution with 4 mL of 14N
246 HNO_3 and 1 mL of 12N HCl . After 1 night on a hot plate, the solution was dried and the residue
247 was dissolved again in 10 ml of 1N HNO_3 . Since filters were not rinsed immediately after
248 filtration with distilled water on board, it was preferable to remove the salt before the column
249 chemistry. Therefore, 40 μL of a Fe solution (60 mg/g) were added. After 1h-heating, Fe was
250 precipitated by raising the pH to about 8 with NH_3 . The Fe precipitate (that coprecipitates Th
251 isotopes) was separated by centrifugation and rinsed several times. Finally, it was dissolved in
252 0.25 ml of 8N HNO_3 , ready for loading on an anionic column. Th isotopes were separated from
253 Fe by ion exchange chromatography on a small volume (0.5 ml) column of AG 1X8 resin (200-
254 400 mesh) with a procedure adapted from Jeandel et al., 2011 (Gdaniec et al., 2018).

255 The total ^{232}Th chemical blank was typically 42 ± 25 pg, although few blanks as high as
256 1400 pg were observed. Given the good agreement between the particulate ^{232}Th concentrations
257 obtained with the Niskin filtrations (see Fig. ES1) and the ISP ^{232}Th concentrations corrected
258 with the low blank values, the highest blanks were not taken into account to correct the ISP
259 data. Total ^{230}Th chemical blank was typically 1 ± 1 fg. As for ^{232}Th , a small number of very
260 large blanks (up to 125 fg) were obtained but assumed to be outliers.

261

262

263 Mass spectrometry

264 For dissolved and in situ pump samples, Th isotopes were measured on a MC-ICPMS
265 Neptune + (Thermo-Fischer). The dissolved samples were analyzed with the initial
266 configuration of the Neptune + with an Apex desolvator. The particulate samples were analyzed
267 after the Neptune's upgrade with a Jet interface and an Aridus 2 desolvating nebulizer. The
268 acquisition conditions are given in Gdaniec et al. (2018). The accuracy of the MC-ICPMS
269 measurement was checked by analysis of Th standard IRMM035 ($^{230}\text{Th}/^{232}\text{Th} =$
270 $(11.31 \pm 0.12) \times 10^{-6}$, $n=6$) and IRMM036 ($(3.059 \pm 0.033) \times 10^{-6}$, $n=8$), in good agreement with
271 reference values: $^{230}\text{Th}/^{232}\text{Th} = (11.38 \pm 0.10) \times 10^{-6}$ for IRMM035 and $^{230}\text{Th}/^{232}\text{Th} =$
272 $(3.047 \pm 0.024) \times 10^{-6}$ for IRMM036 (Sims et al., 2008). The quality of the analyses was checked
273 through the GEOTRACES intercalibration exercise where we obtained precise and accurate
274 dissolved Th concentrations well within the consensus values (Anderson et al., 2012).

275

276 For Niskin filtration samples, ^{232}Th was analyzed by quadrupolar ICPMS (Agilent
277 7500CE of the Observatoire Midi Pyrénées, Toulouse). Indium and rhenium (100 ppt, added to
278 the samples) were used as internal standards to monitor instrument sensitivity variations.

279 Reproducibility calculated from 3 duplicated samples (2 different Niskins) was on average 12%
280 (2 standard deviations). Detection limit was 0.5 pg/kg.

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284 3. Results

285 3.1. Dissolved thorium

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287 Dissolved ^{232}Th concentrations ($^{232}\text{Th}_d$) range from 4.7 pg/kg to 288 pg/kg (Fig. 2a, Tab. ES 1)
288 and generally increase with depth. At stations S1 and S2, the $^{232}\text{Th}_d$ profiles have a local
289 maximum around 200-400 m depth and a sharp increase at bottom depths. Conversely, at station
290 S3, $^{232}\text{Th}_d$ for the deepest sample is relatively low. This feature was also noted for dissolved
291 REE (Garcia-Solsona et al. 2014) and for $^{230}\text{Th}_d$ (see below) analyzed on the same water sample.
292 Along the section, the lowest $^{232}\text{Th}_d$ are found at station S3 and S4, far from the coasts of Africa
293 and Antarctica ($^{232}\text{Th}_d = 4.7\text{-}30$ pg/kg). All the $^{232}\text{Th}_d$ duplicates agree within analytical
294 uncertainties.

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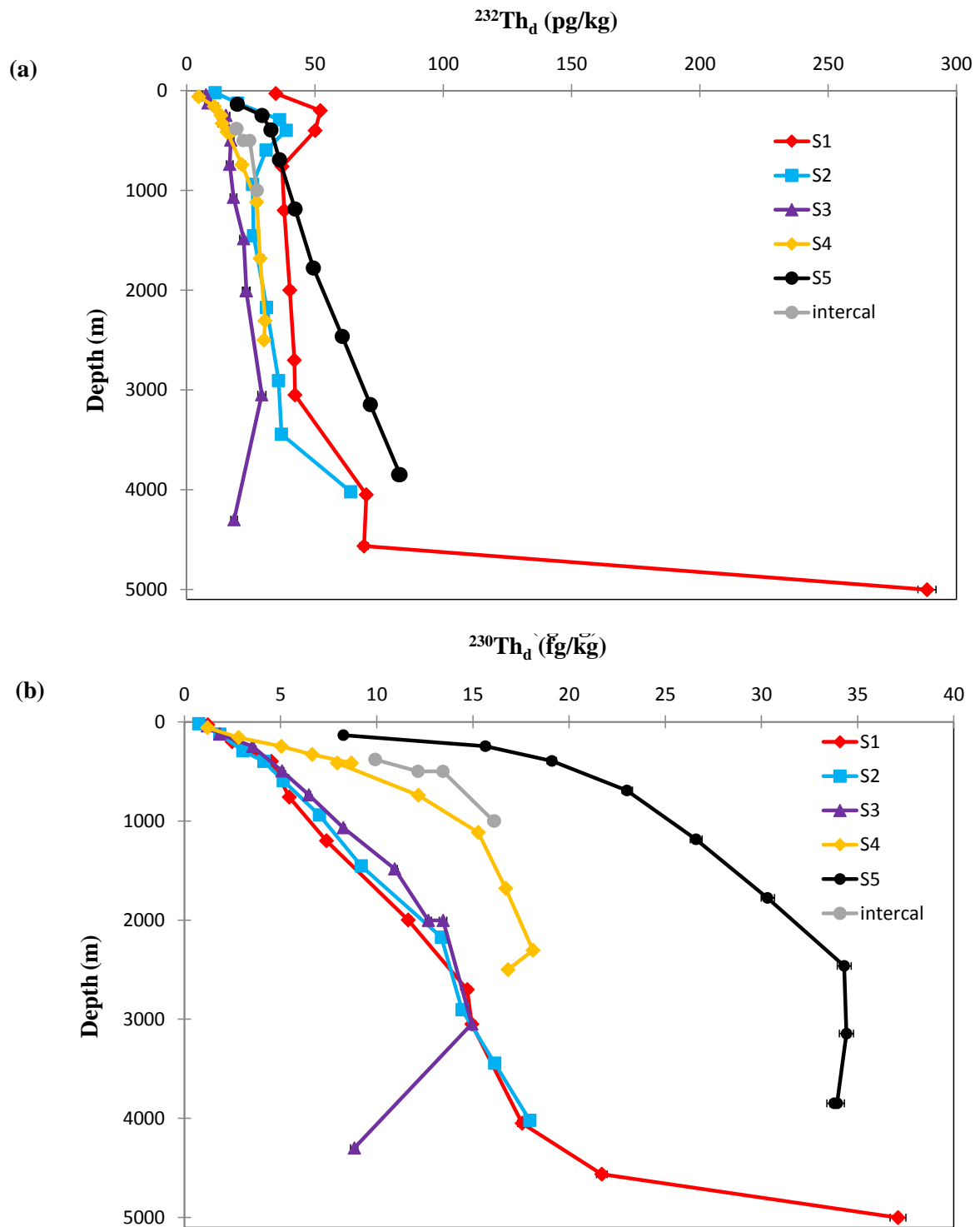


Fig. 3. Dissolved thorium profiles. a: dissolved ^{232}Th . b: dissolved ^{230}Th .

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305 Dissolved ^{230}Th concentrations ($^{230}\text{Th}_d$) range from 0.7 fg/kg to 37 fg/kg (Fig. 3b) and generally
306 increase with depth. At stations S1 and S2, the $^{230}\text{Th}_d$ profiles increase approximately linearly
307 from the surface down to 4000 m depth. At station S1, there is a sharp increase of $^{230}\text{Th}_d$ above
308 the seafloor. Conversely, at station S3, S4 and S5, the $^{230}\text{Th}_d$ profiles have concave shapes and,
309 for a given depth, $^{230}\text{Th}_d$ is higher at stations S4 and S5 than at S1, S2 and S3. All $^{230}\text{Th}_d$
310 duplicates agree within 9% (3 of them agreeing within the analytical uncertainties).

311 Following GEOTRACES good practices, a comparison with 2 stations of the Zero-Drake cruise
312 (Rutgers van de Loeff et al., 2016) is provided (Fig. ES3). Profiles agree within uncertainties
313 for ^{230}Th and more qualitatively for ^{232}Th .

314

315

316 3.2. Particulate thorium

317 Particulate ^{232}Th concentrations ($^{232}\text{Th}_p$) have been already reported elsewhere (Garcia-Solsona
318 et al., 2014). $^{232}\text{Th}_p$ from ISP filtrations range from 0.08 pg/kg to 371 pg/kg (Fig. 4a, Tab. ES2).

319 There is generally a good agreement between $^{232}\text{Th}_p$ profiles obtained with in situ pumps and
320 profiles obtained with Niskin filtrations. The most notable exception is above the seafloor at
321 station S3, where Niskin bottle filtration gives $^{232}\text{Th}_p = 27$ pg/kg, whereas ISP filtration gives
322 $^{232}\text{Th}_p = 2.5$ pg/kg. Apart from this particular feature recorded at S3, the $^{232}\text{Th}_p$ profiles generally
323 increase with depth with a sharp increase above the seafloor. Along the section, the highest
324 $^{232}\text{Th}_p$ concentrations are found at station S1 and S2, which are the stations closest to the African
325 coast.

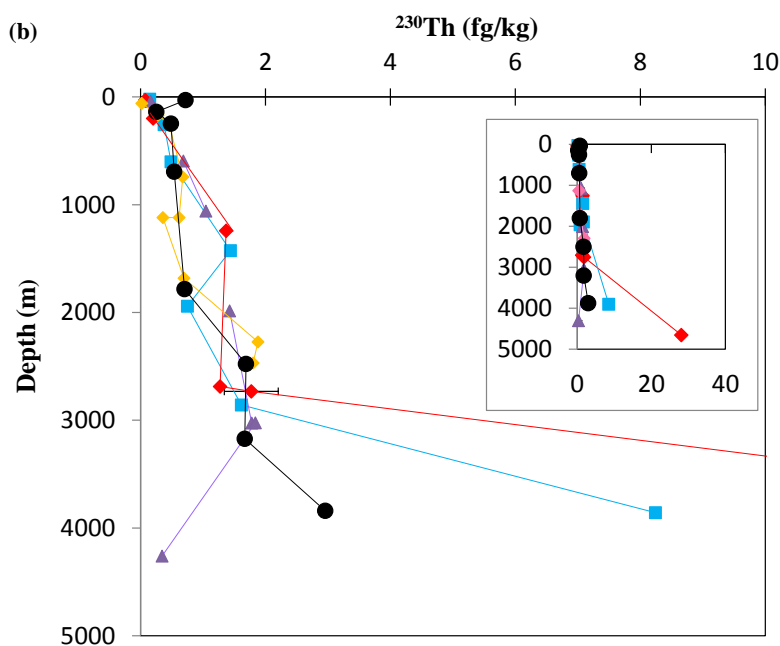
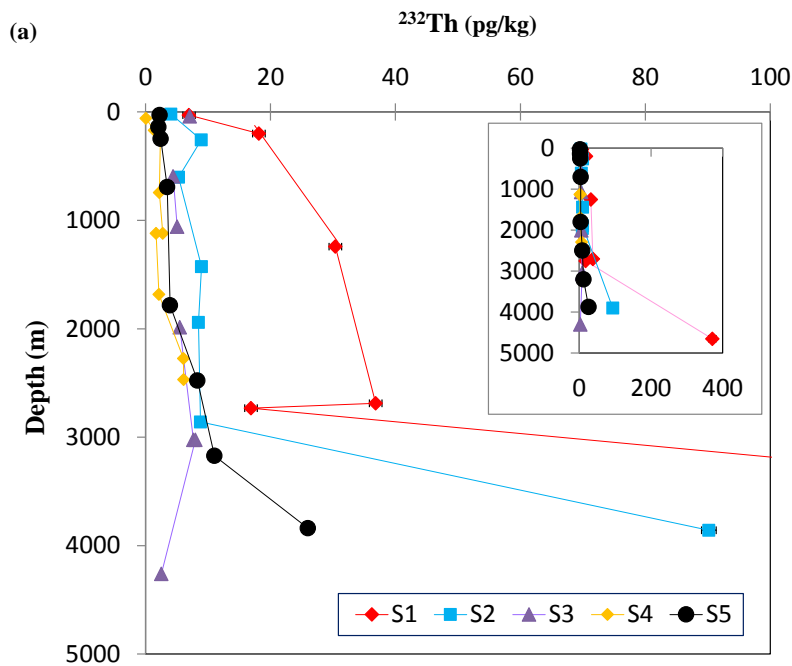
326 All the $^{232}\text{Th}_p$ duplicates agree within analytical uncertainties. The variation in $^{232}\text{Th}_p$ observed
327 between in situ pumps and Niskin filtrations (Fig. ES1) is of the same order than observed for
328 other insoluble elements (Planquette and Sherrell, 2012).

329

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331 Particulate ^{230}Th concentrations ($^{230}\text{Th}_p$) range from 0.02 fg/kg to 28 fg/kg (Fig. 4b). $^{230}\text{Th}_p$
332 generally increase with depth. At stations S1 and S2, the $^{230}\text{Th}_p$ profiles increase approximately
333 linearly from the surface down to 4000 m depth and then present a sharp increase of $^{230}\text{Th}_p$
334 above the seafloor (height above the seafloor is 295 m and 212 m at, respectively, S1 and S2).
335 At station S3, $^{230}\text{Th}_p$ decreases just above the seafloor similarly to $^{232}\text{Th}_d$ (height above the
336 seafloor is 218 m at S3).

337 Two in situ pump filtrations were replicated. At station S3 and 3050-m depth, the
 338 replicates agree within 4% for both $^{232}\text{Th}_p$ and $^{230}\text{Th}_p$. At station S4 and 1130-m depth, both
 339 the $^{232}\text{Th}_p$ and $^{230}\text{Th}_p$ concentrations differ by a factor of ~ 2 , suggesting either a problem of
 340 volume measurement, data recording or a loss of particles during the filter handling. We
 341 choose to use the concentrations measured on the filter with the largest filtered volume of
 342 water, because it corresponds to the $^{232}\text{Th}_p$ concentration closest to the Niskin filtration.
 343
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345

346 Fig. 4. Particulate Thorium profiles collected with *in situ* pumps. (a) Particulate ^{232}Th . (b)
347 Particulate ^{230}Th . Insets: expanded concentration scales to display particulate
348 concentration maximum at S1.

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350

351 4. Discussion

352 4.1. ^{230}Th profiles

353 The dissolved ^{230}Th profiles obtained during Bonus GoodHope are consistent with
354 previous results from the South Atlantic and the Weddell Sea (Rutgers van der Loeff and
355 Berger, 1993, Rutgers van der Loeff et al., 2016). Station S1 has a relatively linear profile from
356 the surface down to 3000 m deep. This is consistent with the 1D reversible scavenging model,
357 in which ^{230}Th is only transported vertically by reversible scavenging on settling particles.
358 However, even though S1 is located in the subtropical gyre, north of the subtropical front, the
359 profile is not perfectly linear. This presumably reflects the influence of surface and intermediate
360 waters advected from the Indian Ocean (ICW and AAIW down to ~1200 m) and deep water
361 advected from the Atlantic Ocean (SE-NADW). The ^{230}Th content of the surface and
362 intermediate waters at S1 compare well with those reported in the south Indian Ocean ($^{230}\text{Th} \approx$
363 4-5 fg/kg at 500 m, Thomas et al., 2006). The ^{230}Th contents of deep waters ($^{230}\text{Th} \approx 13$ fg/kg
364 at 2000 m) at S1 are on the low side of those reported for SE-NADW sampled off Namibia
365 ($^{230}\text{Th} \approx 12$ -24 fg/kg at 2000 m, Scholten et al. 2008) and that flows towards S1 (Arhan et al.,
366 2003), suggesting the imprint of boundary scavenging. The deepest parts of the S1 profile
367 significantly deviate from the linear increase: the higher dissolved and particulate ^{230}Th (as well
368 as ^{232}Th) concentrations suggest a release from the South African margin sediments.

369 From the surface down to 3000 m depth, the ^{230}Th profiles of S2 and S3 are similar to
370 S1. In the deepest waters, S2 has a strong increase of its dissolved and particulate ^{230}Th
371 (although less than S1) also suggesting inputs from the sediments or lateral transport from the
372 African margin. By contrast, at S3, the low $^{230}\text{Th}_d$ concentrations at a depth of 4500 m could
373 also be attributed to bottom scavenging (a low REE content on the same sample was also noted
374 by Garcia-Solsona et al., 2014).

375 The strongly concave shapes of the $^{230}\text{Th}_d$ profiles at S4 and S5 strongly suggests these
376 profiles are impacted by the upwelling of LCDW (Rutgers van der Loeff and Berger, 1993).
377 The $^{230}\text{Th}_d$ profile at S3 has also a concave shape that can be regarded as a result of the LCDW
378 upwelling. However, above 3000 m depth, it is not so different from the S1 profile where no

379 LCDW upwelling is suspected. Also, the low $^{230}\text{Th}_d$ concentrations at a depth of 4500 m, that
380 accentuate the nonlinearity of the profile, could also be attributed to bottom scavenging.

381

382 At stations S1, S2 and S3, the $^{230}\text{Th}_p$ profiles have very similar slopes, particularly from the
383 surface down to 1500 m depth. We consider the conservation equation of the 1D reversible
384 scavenging model at steady state (and neglecting ^{230}Th radioactive decay):

$$385 \quad \frac{\partial C_t}{\partial t} = P_d - w_p \frac{\partial C_p}{\partial z} = 0 \quad (2)$$

386 Where C_t is the total (dissolved + particulate) ^{230}Th concentration, P_d is the in situ production
387 rate of ^{230}Th ($P_d = 0.56 \text{ fg/kg/y}$), w_p is the particle settling speed and C_p the particulate ^{230}Th
388 concentration. Hence, w_p is given by: $w_p = P_d / (\partial^{230}\text{Th}_p / \partial z)$. Using an average slope of 0.001
389 fg/kg/m, we obtain that $w_p = 560 \text{ m/d}$, in agreement with Rutgers van der Loeff and Berger
390 (1993).

391

392

393 In the southern part of the section, we first use the mixing-scavenging model
394 introduced by Rutgers van der Loeff and Berger (1993) to evaluate the impact of lower
395 Circumpolar Deep Water upwelling and input into the Weddell Gyre. This “scavenging-
396 mixing” model relies on several assumptions:

397 (i) The representation of mixing with a constant restoring term at all depths (eq. 3). This is
398 largely ad hoc, so that the physical interpretation of C_{it} and τ_w is unclear.

399 (ii) the omission of the effects of vertical transport by advection and diffusion,

400 (iii) steady state,

401 (iv) the partition coefficient K is vertically uniform. It implies that the $^{230}\text{Th}_{xs}$ particulate
402 fraction is constant over the depth profile. Clear variations of the $^{230}\text{Th}_{xs}$ particulate fraction
403 (Tab. ES2) occur in the deepest samples either due to particle inputs from the margins
404 (stations S1, S2 and S5, see Gdaniec et al., 2019) or following hydrothermal scavenging
405 (station S3).

406 (v) K is very small compared to 1, so that C_d could be approximated by C_t . Apart for the
407 deepest samples at stations S1 and S2, it introduces an underestimation of w_p by 13 % at most.

408 (vi) the particle settling velocity w_p is vertically uniform. This assumption is deduced from
409 linear dissolved and particulate ^{230}Th profiles observed in regions unaffected by strong deep
410 currents (Roy-Barman et al., 1996).

411 (vii) C_d is negligible near the surface.

412

413 Using the above assumptions, the tracer conservation equation of this model is:

414

$$415 \quad \frac{\partial C_t}{\partial t} = P_d + \frac{(C_{i,t} - C_t)}{\tau_w} - w_p \frac{\partial C_p}{\partial z} = 0 \quad (3)$$

416 Where τ_w is restoring time scale taken equivalent to the water residence time and $C_{i,t}$ is
417 total ^{230}Th concentration of the inflowing water. Introducing a partition coefficient between
418 dissolved and particulate Th, $K = C_p / C_d$ and integrating equation 3 vertically over the water
419 column, the $^{230}\text{Th}_d$ concentration at any depth is given by:

$$420 \quad C_d = (C_{i,t} + P_d \tau_w) \times \left(1 - \exp\left(-\frac{z}{w_p K \tau_s}\right) \right) \quad (4)$$

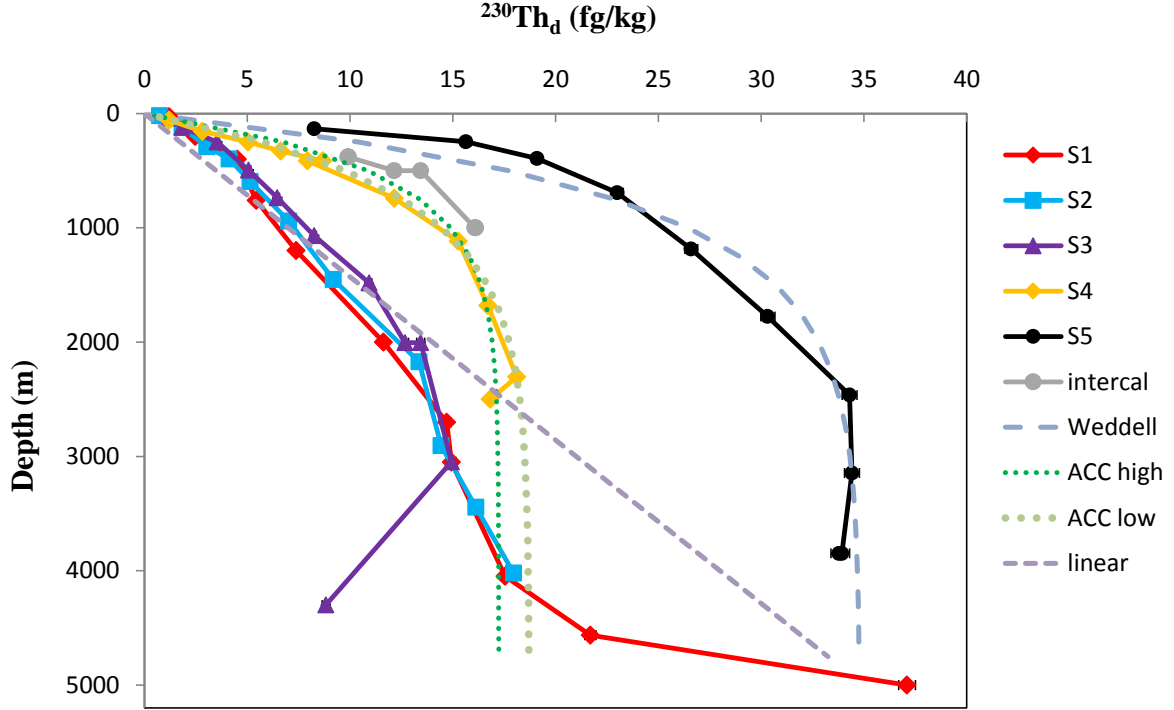
421

422 For station 5, we use the $^{230}\text{Th}_i = 24$ fg/kg for the LCDW (Rutgers van der Loeff and Berger,
423 1993), $K=0.04$ is the average ratio measured at this station. Then, we adjust $w_p = 875$ m/y and
424 $\tau_w = 20$ y to obtain a good agreement between the modelled curve and the data (Fig. 5). The
425 low τ_w obtained here compared to 45 y used by Rutgers van der Loeff and Berger (1993)
426 derives presumably from the relatively peripheral position of station S5 in the Weddell Sea.
427 We confirm that the scavenging residence time in the Weddell Sea ($\tau_s = d / (2Kw_p) = 55$ y for a
428 water column of a depth of $d = 4000$ m) is larger than in the subtropical gyre ($\tau_w = 25$ y for a
429 water column of a depth of 4000 m). On the whole, the model explains why the $^{230}\text{Th}_d$
430 concentrations are high and do not increase linearly with depth in the Weddell Gyre.

431 We apply the same model to station S4. Here $^{230}\text{Th}_i$ is also adjusted to obtain a good
432 data-model agreement. We note that 2 modeled curves based on relatively different sets of τ_w
433 and w_p values (ACC high and ACC low, Fig. 5) agree with the S4 data. This illustrates a limit
434 of this method to estimate particle settling velocities.

435

436



437
 438 Fig. 5. Modelling of the Bonus Good Hope profiles. The dotted and dashed curves are calculated with
 439 Eq. 4. Weddell: $C_i = 24$ fg/kg, $w_p = 875$ m/y, $\tau_w = 20$ y and $K = 0.04$. ACC low: $C_i = 12$ fg/kg, $w_p =$
 440 803 m/y, $\tau_w = 12$ y and $K = 0.07$. ACC high: $C_i = 15$ fg/kg, $w_p = 1785$ m/y, $\tau_w = 4$ y and $K = 0.125$.
 441 linear: $C_i = 0$ fg/kg, $w_p = 800$ m/y, $\tau_w = +\infty$ and $K = 0.1$.

442
 443

444 This advection-scavenging model has been largely used to describe the vertical
 445 distribution of ^{230}Th in the Southern Ocean (Coppola et al., 2006, Venchiarutti et al., 2011,
 446 Rutgers van der Loeff et al., 2016). Despite crude assumptions (e.g., uniform influence
 447 throughout the water column by a single water mass), this mixing-scavenging model has the
 448 great merit of highlighting the impact of upwelling on $^{230}\text{Th}_d$ profiles and to provide an
 449 estimate of the particle settling speed when the 1D production-scavenging model cannot be
 450 used. Nevertheless, this model cannot fully describe the upwelling and mixing of 2 different
 451 water masses along an isopycnal surface as observed during BGH and described in the next
 452 section. Salinity data help to illustrate this point. For a conservative tracer ($P_d = 0$, $w_p = 0$ and
 453 $C_p = 0$) and assuming steady state, equation (3) reduces to:

$$454 \quad (S_{t_i} - S_t) = 0 \quad (5)$$

455 This expresses that salinity is constant and equal to the salinity of the inflowing water ($S_t = S_{t_i}$).
 456 In this model, LCDW is advected at all levels in the Weddell Gyre. It implies that the Weddell
 457 Gyre must be filled only with LCDW. Nevertheless, the $^{230}\text{Th}_t$ concentration in the Weddell

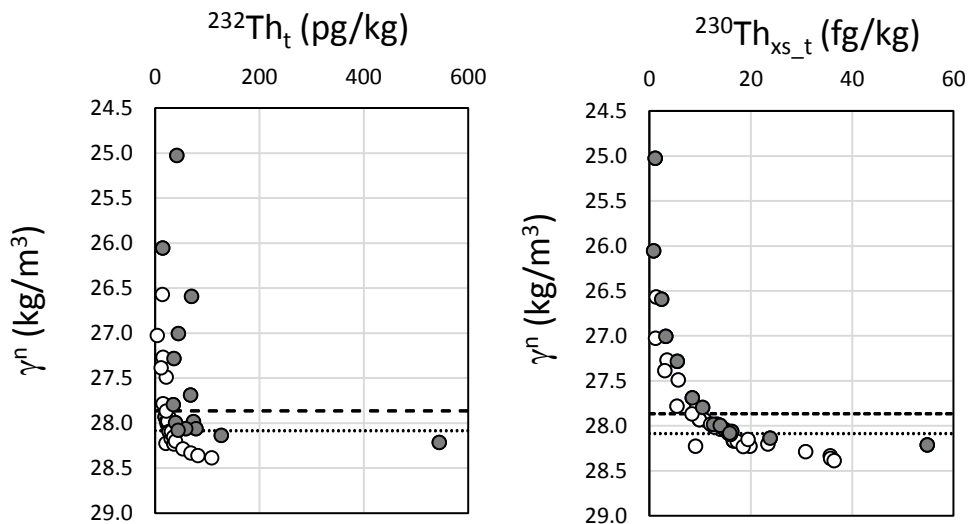
458 Gyre changes with depth due to the scavenging transport of $^{230}\text{Th}_t$ by sinking particles. Hence,
 459 this mixing-scavenging model cannot represent the mixing of water masses with different
 460 origins and labeled with different θ and S signatures as they are observed along the BGH section
 461 and elsewhere in the Southern Ocean. In principle, general circulation models, associated or
 462 coupled with biogeochemical model, have the potential to reproduce the coupled effect of
 463 scavenging and mixing, but the present generation of coupled models do not accurately describe
 464 the vertical distribution of scavenging intensity (e.g.: Dutay et al., 2009, Gu and Liu, 2017, Van
 465 Hulten et al., 2018).

466

467 4.2. Th isotopes along neutral density surfaces

468 While there are large horizontal concentration gradients of $^{230}\text{Th}_d$ and total (dissolved +
 469 particulate) ^{230}Th ($^{230}\text{Th}_t$) along the BGH section (mostly between station S3 to S5, Fig 3),
 470 $^{230}\text{Th}_t$ concentration gradients are small on a given neutral density surface (Fig. 6). Figure 6
 471 highlights that distribution of $^{230}\text{Th}_d$ is mainly driven by transport from upwelling and mixing
 472 along isopycnal surfaces. There is more variability of $^{232}\text{Th}_d$ or $^{232}\text{Th}_t$ (dissolved + particulate
 473 ^{232}Th) on a given neutral density value, presumably because large lithogenic inputs occur at
 474 stations S1 and S2 (Fig. 6).

475



476

477 Fig. 6. Neutral density versus $^{232}\text{Th}_t$ and $^{230}\text{Th}_t$ concentration. Grey dots: S1 and S2 stations
 478 with high $^{232}\text{Th}_t$ contents. White dots: S3, S4 and S5. Dashed line: $\gamma^n = 27.865 \text{ kg/m}^3$. Dotted
 479 line: $\gamma^n = 28.094 \text{ kg/m}^3$.

480

481

482 In the following, we discuss concentration changes along isopycnal surfaces. As these
483 changes are small, we restrict this discussion to the Bonus GoodHope data, all generated in the
484 same laboratory, in order to avoid the systematic inter-laboratory differences identified during
485 the GEOTRACES intercalibration (Anderson et al., 2012).

486 We focus on 2 neutral density surfaces: $\gamma^n = 27.865 \text{ kg/m}^3$ and $\gamma^n = 28.094 \text{ kg/m}^3$ (Fig
487 ES2). The neutral density surface $\gamma^n = 27.865 \text{ kg/m}^3$ can be followed from $z=1500 \text{ m}$ at station
488 S1 to $z=139 \text{ m}$ at station S5. It crosses the following water masses: I-UCDW (S1), A-UCDW
489 (S2 and S3), DP-UCDW (S4) and AASW/WW (S5). The neutral density surface: $\gamma^n = 28.094$
490 kg/m^3 can be followed from $z = 3000 \text{ m}$ at station S1 to $z = 200 \text{ m}$ at station S5. It is close to
491 the neutral density of the deep salinity maximum observed in 2004 along the same section
492 (Gladyshev et al., 2008). It crosses the following water masses: SE-NADW (S1 and S2), SW-
493 NADW/AABW (S3), LCDW (S4) and AASW/WW (S5).

494
495 We choose these two neutral density surfaces because: (1) they are found along the whole
496 section, (2) samples along these surfaces tend to form a linear trend on a θ -S diagram, indicating
497 a binary mixing (Fig. 7), and (3) they are not interrupted by the Agulhas Ridge. When, at a
498 given station, no sample was collected exactly on the selected neutral density surface,
499 concentrations were linearly interpolated between the samples collected just above and below
500 this surface at the same station. When the particulate Th was not analyzed with *in situ* pump
501 samples at the target depth or in its vicinity (e.g., intercal station), we used the $^{232}\text{Th}_p$
502 concentration obtained with the Niskin filtration profiles and the $^{230}\text{Th}_p$ was calculated with the
503 $(^{230}\text{Th}_p/^{232}\text{Th}_p)$ ratio linearly interpolated between the ratios measured on particles just above
504 and below or at the nearby station:

$$505 \quad ^{230}\text{Th}_{p\text{-estimates}} = ^{232}\text{Th}_{p\text{-Niskin filtration}} * (^{230}\text{Th}_p/^{232}\text{Th}_p)_{\text{interpolated}} \quad (6)$$

506

$$507 \quad \underline{\gamma^n = 27.865 \text{ kg/m}^3}$$

508 On a θ -S diagram, S1, S2, S3 and S4 plot along a straight line, suggesting a binary mixing
509 between S1 and S4 (Fig. 7a). S5 does not fall on this line, possibly due to ocean-atmosphere
510 interactions (Abadie et al., 2017), so that this station does not correspond to the same binary
511 mixing along this neutral density surface. Therefore, in the following, we will apply the mixing
512 model mostly between stations S1 and S4 for this isopycnal.

513 On a S- $^{232}\text{Th}_t$ diagram, the stations S2 and S3 stand below the conservative mixing line between
514 S1 and S4 (Fig. 7c). It corresponds to a depletion (net removal of ^{232}Th on sinking particles) of

515 $^{232}\text{Th}_t$ compared to a conservative mixing between S1 and S4: $\Delta^{232}\text{Th}_t < 0$. The strong $^{232}\text{Th}_t$
516 inputs from the South African margin to the ACC are marked by a strong $^{232}\text{Th}_t$ concentration
517 gradient ($^{232}\text{Th}_t$ decrease by a factor ~ 4 between stations S1 and S4). A small $^{232}\text{Th}_t$ gradient
518 between Station S5 and S4 suggests smaller inputs from the Weddell Sea.

519

520 On a S- $^{230}\text{Th}_t$ diagrams, there is much less change in $^{230}\text{Th}_t$ concentration ($\sim 30\%$ between
521 stations S1 and S4) compared to $^{232}\text{Th}_t$ presumably because the margin is not a strong source of
522 $^{230}\text{Th}_t$ for the ACC. Nevertheless, the $^{230}\text{Th}_t$ concentrations generally decrease southward.
523 However, stations S2 and S3 stand above the conservative mixing line between S1 and S4 (Fig.
524 7e). This trend would correspond to an excess of $^{230}\text{Th}_t$ compared to a conservative mixing
525 between S1 and S4: $\Delta^{230}\text{Th}_t > 0$. There is an accumulation of ^{230}Th due to *in situ* production
526 despite the net removal of $^{232}\text{Th}_t$ during the same mixing process.

527

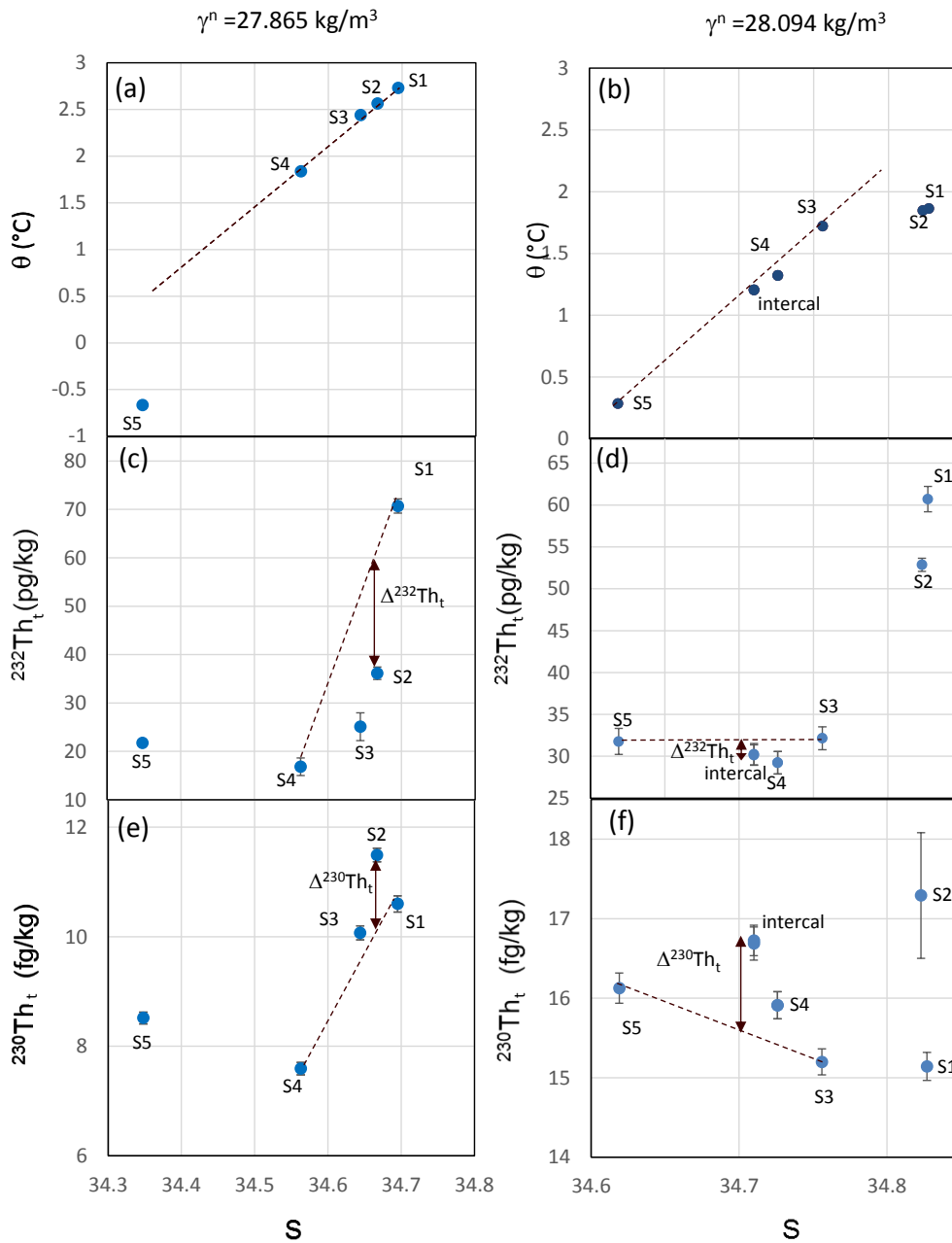
528 $\gamma^n = 28.094 \text{ kg/m}^3$

529 On a θ -S diagram, S3, S4, intercal and S5 plot along a straight line, suggesting a binary
530 mixing between S3 and S5 (Fig. 7b). S1 and S2 do not fall on this line, so that these 2 stations
531 do not correspond to the same binary mixing along this neutral density surface. Therefore, in
532 the following, we will restrict our analysis to stations S3 to S5 and, in the next section, we will
533 apply the mixing model only between stations S3 and S5.

534 On a S- $^{232}\text{Th}_t$ diagram, we observe the same trend as on the previous isopycnal: a strong
535 $^{232}\text{Th}_t$ decrease from S1 to S3 and a weaker decrease from S5 to S4 (Fig 7d). This illustrates the
536 role of the margins (and particularly the South African margin) as sources of ^{232}Th for the ACC.
537 We argued in the previous paragraph that waters from stations S1 and S2 do not seem directly
538 involved in the mixing observed further south. However, high $^{232}\text{Th}_t$ concentrations are also
539 observed in the SW-NADW sampled upstream of the BGH section. These high values are
540 acquired as this water flows along the Namibian margin (Scholten et al., 2008) so that it could
541 be a suitable $^{232}\text{Th}_t$ source fueling the southern part of the BGH section. S4 and intercal stations
542 stand below the S3-S5 binary mixing line, reflecting $^{232}\text{Th}_t$ depletion at these 2 stations
543 compared to conservative mixing, expressed as $\Delta^{232}\text{Th}_t < 0$ (Fig 7). More generally, paired
544 $^{232}\text{Th}_t$ -S data indicates that there is a net Th removal by the settling particles while water masses
545 mix between stations S3 and S5.

546

547 As for $\gamma^n = 27.865 \text{ kg/m}^3$, the relative variations of $^{230}\text{Th}_t$ are small (Fig. 7f). On a S-
 548 $^{230}\text{Th}_t$ diagram, the S4 and intercal stations stand above the S3-S5 binary conservative mixing
 549 trend (S1 and S2 are not considered because they are not involved in the mixing on this
 550 isopycnal). This trend would correspond to an excess of $^{230}\text{Th}_t$ compared to a conservative
 551 mixing: $\Delta^{230}\text{Th}_t > 0$. Like on the previous isopycnal, ^{230}Th accumulates due to *in situ* production
 552 despite the net removal of $^{232}\text{Th}_t$ during water mass mixing.



553
 554 Figure 7: Potential temperature, $^{232}\text{Th}_t$ and $^{230}\text{Th}_t$ versus salinity on different neutral density surfaces.
 555 Left: $\gamma^n = 27.865 \text{ kg/m}^3$. Right: $\gamma^n = 28.094 \text{ kg/m}^3$. a-b: Potential temperature versus salinity. c-d: $^{232}\text{Th}_t$

556 versus salinity. e-f: $^{230}\text{Th}_t$ versus salinity. Thin dashed line: conservative binary mixing line. Vertical
557 double arrows denote the deviation compared to conservative mixing.

558 See table ES3 for the data calculated on isopycnals.

559

560 4.3. Modeling advection-diffusion-scavenging along a neutral density surface.

561 In the deep ocean, water mass mixing is thought to occur preferentially along
562 isopycnal surfaces. There is both advection and eddy diffusion along the steep isopycnal
563 surfaces of the Southern Ocean (Zika et al., 2009, Naveira Garabato et al., 2007). While
564 diapycnal mixing is important in the Drake Passage due to the rough topography, it appears to
565 be much weaker out of the passage (Watson et al., 2013), so we will neglect it for the present
566 analysis. We approximate the combined effect of advection and diffusion along an isopycnal
567 by extending the conservation equation (2) with the explicit introduction of advection and
568 diffusion terms in an infinitesimal volume of water at steady state (Jenkins, 1987):

$$569 \quad K_i \frac{\partial^2 C_t}{\partial x^2} - u_i \frac{\partial C_t}{\partial x} + P_d - w_p \left(\frac{\partial C_p}{\partial z} \right) = 0 \quad (7)$$

570 Where C_t is the total (dissolved + particulate) concentration of the tracer, C_p the particulate
571 concentration of the tracer, K_i is the cross-stream isopycnal eddy diffusion coefficient and u_i
572 is the cross-stream isopycnal advection velocity. The x-axis is parallel to the isopycnal surface
573 in the meridional direction and oriented northward. The z axis is oriented perpendicular to the
574 isopycnal surface and hence is almost vertical. Just like in the advection-scavenging model
575 (equ. 3), keeping w_p out of the partial derivative with respect to z (writing $w_p \left(\frac{\partial C_p}{\partial z} \right)$ instead of
576 $\left(\frac{\partial (w_p C_p)}{\partial z} \right)$) requires to assume that w_p does not vary with z. We integrate Eq. 7 along
577 isopycnal surfaces of the BGH section between the stations corresponding to the end members
578 of water mass mixing (stations for S1 and S4 for $\gamma^n = 27.865 \text{ kg/m}^3$ and S3 and S5 for γ^n
579 $= 28.094 \text{ kg/m}^3$). Unlike most Th models that are built by vertically integrating the
580 conservation equation, here we integrate the conservation equation along surfaces that are
581 only slightly tilted relative to the horizontal. This is a simplified view of water transport
582 because while water moves across the ACC, it is also rapidly advected eastward by the ACC
583 (S2-S4) and the northern limb of the Weddell Gyre (S5). We assume that K_i , u_i , w_p and
584 (dC_p/dz) are all constant along x in order to obtain analytical solutions. We note $C_t = C_{t\text{-conc}} +$
585 ΔC_t , where $C_{t\text{-conc}}$ is the concentration of the tracer if it had a conservative behavior and ΔC_t
586 (as plotted in Fig 7) is the deviation of this tracer compared to a conservative behavior. The
587 solution is then:

588

$$589 \quad C_{t-conc} = \frac{\frac{u_i}{K_i}x - e^{\frac{u_i}{K_i}x_B}}{\frac{u_i}{K_i}x_A - e^{\frac{u_i}{K_i}x_B}} (C_{t_A} - C_{t_B}) + C_{t_B} \quad (8)$$

$$590 \quad \Delta C_t = \frac{(P_d - w_p \frac{dC_p}{dz})}{u} \left((x - x_B) + (x_B - x_A) \left(\frac{\frac{u_i}{K_i}x - e^{\frac{u_i}{K_i}x_B}}{\frac{u_i}{K_i}x_A - e^{\frac{u_i}{K_i}x_B}} \right) \right) \quad (9)$$

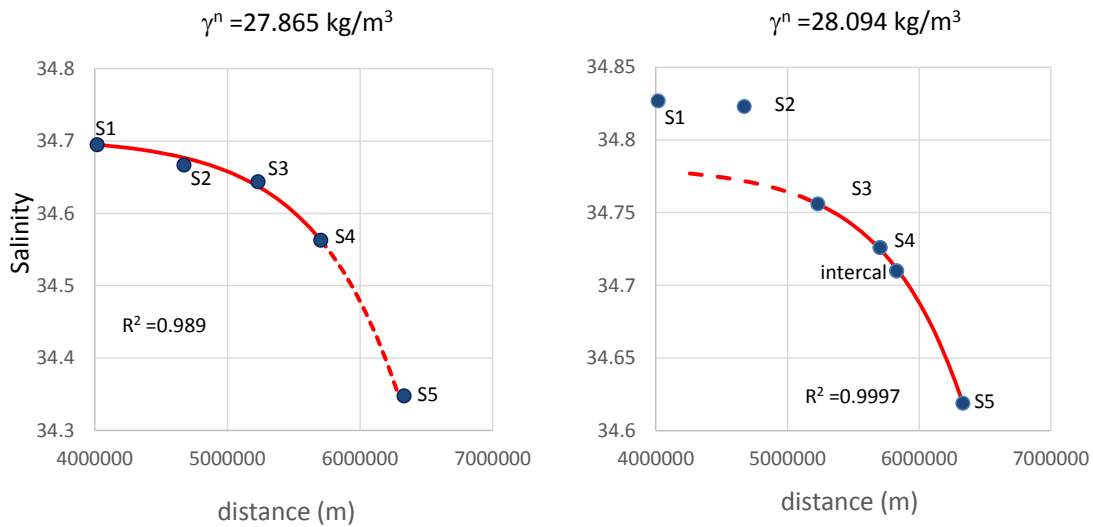
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592

593 Here A and B represent stations S1 and S4 or S3 and S5 along the neutral density surface. For
 594 a conservative tracer such as salinity, the concentration is given by:

$$595 \quad S = \frac{e^{\frac{u_i}{K_i}(x-x_A)} - 1}{e^{\frac{u_i}{K_i}(x_B-x_A)} - 1} (S_B - S_A) + S_A \quad (10)$$

596



597

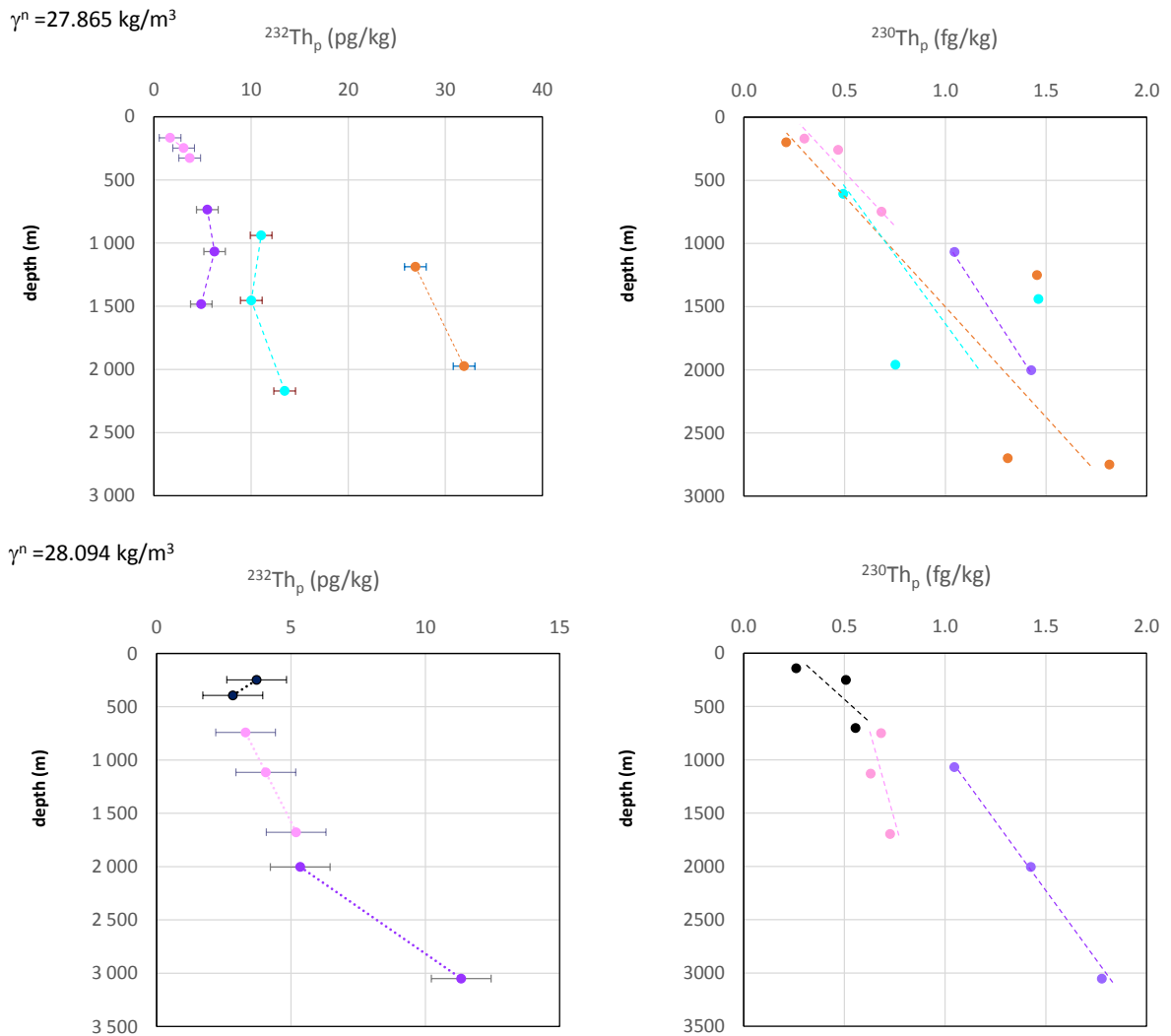
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599 Figure 8: Advection-diffusion modelling of salinity. The distance considered is the meridian distance
 600 from the equator ($d = 0$ at the equator). Red curves are obtained with equation 10. u_i/K_i is adjusted by
 601 least squares just based on the stations corresponding to the 2-water mass mixing and by forcing the
 602 curves to pass through the 2 water mass end members. The values of u_i/K_i are given in Table 1. Dotted
 603 curves are extrapolated to the rest of the section but the corresponding stations are not used to calculate
 604 the curve parameters.

605

606

607



609

610 Figure 9: $^{232}\text{Th}_p$ and $^{230}\text{Th}_p$ profiles. For each isopycnal, we also plot the sample analyzed just above
 611 and below in order to define the slopes ($d^{232}\text{Th}_p/dz$) and ($d^{230}\text{Th}_p/dz$), respectively. Color code is the
 612 same as in Figure 3.

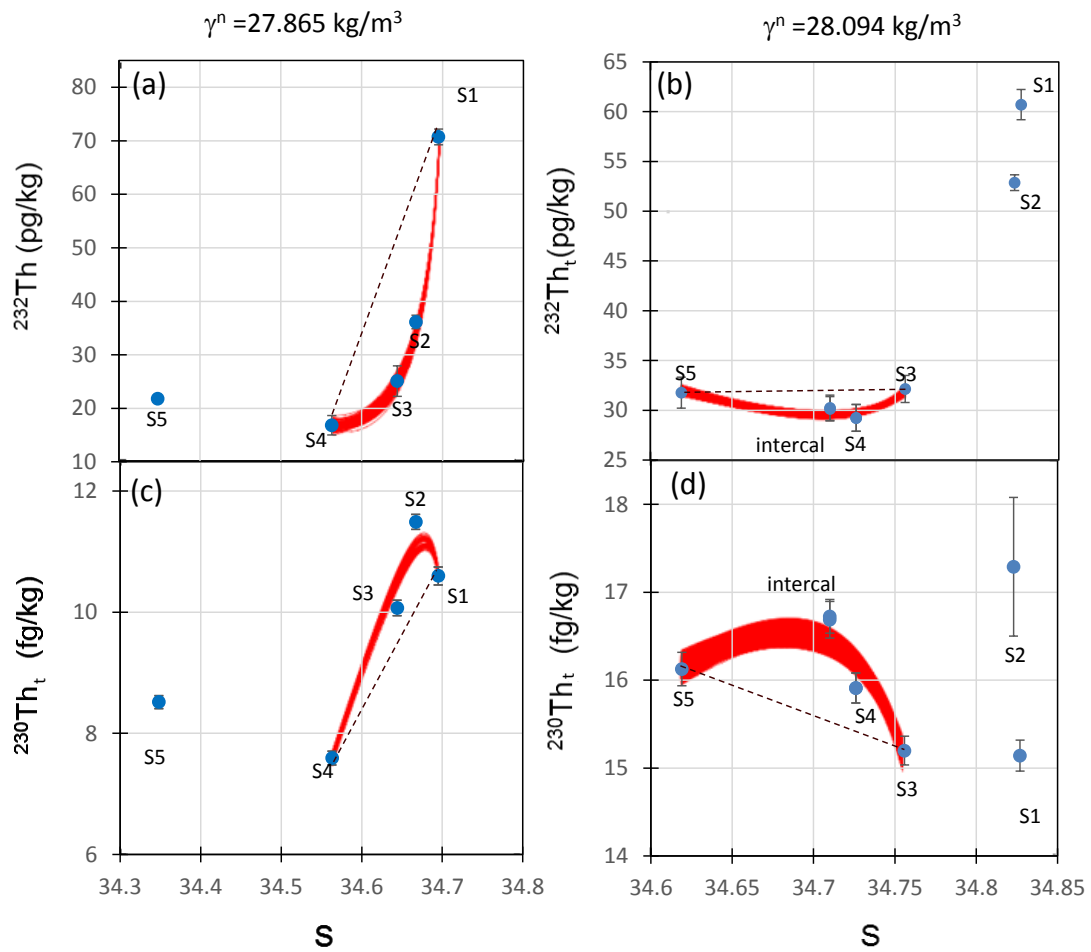
613

614

615 $\gamma^n = 27.865 \text{ kg/m}^3$

616 Equation 10 allows determining the u_i/K_i ratio using salinity data along the BGH
 617 section. For $\gamma^n = 27.865 \text{ kg/m}^3$, we obtain $u_i/K_i = -1.57 \times 10^{-6} \text{ m}^{-1}$ (Fig. 8, Tab. 1). The minus
 618 sign corresponds to a southward advection ($u_i < 0$ and K_i is always positive). This is consistent
 619 with the southward upwelling expected based on the average circulation of UCDW in the
 620 Southern Ocean within this density range (section 2.1). This feature is observed for the 2
 621 isopycnals studied here (see below for $\gamma^n = 28.094 \text{ kg/m}^3$).

622



624

625 Figure 10: Advection-diffusion modelling of S and Th_t isotopes compared to data. Staked red
 626 curves: modelled curves with the highest R^2 values (Tab. 1). Thin black dotted line:
 627 conservative mixing between S3 and S5.

628

629

630 Tab. 1: Model parameters and outputs.

	$d^{232}\text{Th}_p/dz$ (pg/kg/m)	$d^{230}\text{Th}_p/dz$ (fg/kg/m)	u_i/K_i (m^{-1})	K_i (m^2/s)	u_i (m/s)	w_p (m/y)	R^2_{-0}	R^2_{-2}
$\gamma^n = 27.865$ kg/ m^3	- 0.001 / + 0.012	$3.8 \times 10^{-4} - 6.2 \times 10^{-4}$	-1.57×10^{-6}	2000 ± 840	- 0.0033 ± 0.0014	674 ± 250	~ 0.945	~ 0.994
$\gamma^n = 28.094$ kg/ m^3	+ 0.001 / + 0.009	$0.4 \times 10^{-4} - 3.5 \times 10^{-4}$	-1.65×10^{-6}	2180 ± 480	- 0.0036 ± 0.0008	418 ± 470	~ 0.806	~ 0.696

631

632 To determine $d^{232}\text{Th}_p/dz$, we use the Niskin filtration data because they have a better vertical
 633 resolution. We determine $d^{232}\text{Th}_p/dz$ based on the $^{232}\text{Th}_p$ concentrations measured above and

634 below the depth of the neutral density surface at each station (Fig. 9). For $\gamma^n = 27.865 \text{ kg/m}^3$,
635 $d^{232}\text{Th}_p/dz$ varies strongly between S1 and S4. Interestingly, we obtain low $d^{232}\text{Th}_p/dz$ values
636 when the $^{232}\text{Th}_t$ horizontal gradient is low (e.g., $d^{232}\text{Th}_p/dz = - 0.001 \text{ pg/kg/m}$ at S3) and
637 higher $d^{232}\text{Th}_p/dz$ values when the $^{232}\text{Th}_t$ horizontal gradient is high (e.g., $d^{232}\text{Th}_p/dz = +$
638 0.006 pg/kg/m at S1). In other words, the increase of $^{232}\text{Th}_t$ with depth is due to the horizontal
639 transport of ^{232}Th by eddy diffusion from the margin, whereas without inputs from the
640 margins the dissolved and particulate ^{232}Th concentration would be expected to be constant
641 with depth (Roy-Barman et al., 1996). Ideally, it would be best to integrate equation 7 with
642 $d^{232}\text{Th}_p/dz$ and $d^{230}\text{Th}_p/dz$ varying with x . In practice, it is difficult to determine the evolution
643 of these gradients along the isopycnal or even to determine the exact average gradients
644 between stations S1 and S4. As a first approach, we will just use the range of the gradients
645 measured between S1 and S4 as input values to determine w_p , u_i and K_i . For ^{232}Th , we use a
646 range $d^{232}\text{Th}_p/dz$ from $- 0.001$ to $+ 0.012 \text{ pg/kg/m}$ (Fig. 9, Tab. 1). For ^{230}Th , most $^{230}\text{Th}_p$ data
647 plot along a slope ranging between $3.8 \times 10^{-4} \text{ fg/kg/m}$ and $6.2 \times 10^{-4} \text{ fg/kg/m}$ (Fig. 9, Tab. 1).

648 We then estimate w_p and u_i with a least square approach. Using Monte Carlo
649 simulations, we maximize the coefficient of determination (R^2) of the $^{230}\text{Th}_t$ versus S and
650 $^{232}\text{Th}_t$ versus S curves simultaneously. In equation 9, w_p and the $d\text{Th}_p/dz$ terms are present as
651 products so that their opposite variations cancel each other. As a consequence, a relatively
652 large range of w_p , $d^{230}\text{Th}_p/dz$ and $d^{232}\text{Th}_p/dz$ values yield similar R^2 values. Therefore, we
653 report w_p and u_i obtained for a range of high R^2 values. We obtain $w_p = 674 \pm 250 \text{ m/y}$ and u_i
654 $= - 0.0033 \pm 0.0014 \text{ m/s}$. Using the u_i/K_i ratio estimated previously, we deduce $K_i = 2000 \pm$
655 840 m/s^2 .

656

657 $\gamma^n = 28.094 \text{ kg/m}^3$

658 For $\gamma^n = 28.094 \text{ kg/m}^3$, most of the $^{230}\text{Th}_p$ data plot along a slope of $5 \times 10^{-4} \text{ fg/kg/m}$,
659 whereas $^{232}\text{Th}_p$ data plot along a slope of $2 \times 10^{-4} \text{ pg/kg/m}$ (Fig. 10). In both diagrams, the
660 slope of the S4 station is poorly defined and potentially lower than the average. Using the
661 same method, we obtain $u_i/K_i = - 1.65 \times 10^{-6} \text{ m}^{-1}$, $w_p = 435 \pm 324 \text{ m/y}$, $u_i = - 0.0036 \pm 0.0008$
662 m/s and $K_i = 2180 \pm 480 \text{ m/s}^2$ (Fig 8-9, Tab. 1).

663

664 The 2 K_i values estimated here are on the upper end of the range (300 to 1800 m^2/s) reported
665 in the Southern Ocean, whereas u_i is on the lower but of the same order of magnitude than an

666 estimate of the cross-stream horizontal velocity associated with the upwelling in the ACC (~ -
667 0.010 ± 0.003 m/s, Naveira Garabato et al., 2007, Zika et al., 2009).

668
669 We have estimated w_p at relatively close depths on the 2 isopycnals (1200-1500 m for $\gamma^n =$
670 27.865 kg/m³ at stations S3 and S2 and 1000-1200 m for $\gamma^n = 28.094$ kg/m³ at stations intercal
671 and S4, Fig. ES2). Taking the corresponding w_p at face value (but keeping in mind that they
672 strongly rely on the particulate concentration gradient estimate), it seems that there is a
673 southward decrease of w_p (for $\gamma^n = 27.865$, $w_p = 600$ m/y and for $\gamma^n = 28.094$ kg/m³, $w_p = 300$
674 m/y). This southward decrease of settling speed seems consistent with the ²³⁸U-²³⁴Th
675 disequilibrium in the surface waters that decreased from the north to the south of the polar
676 front zone (PFZ) during the BGH cruise (Planchon et al., 2013) and 2 weeks earlier on the
677 same transect during the ANTXIV/3 cruise (Rutgers van der Loeff et al., 2011). This
678 gradient of the ²³⁸U-²³⁴Th disequilibrium indicates a higher removal rate of ²³⁴Th and suggests
679 a higher particle settling velocity in the north compared the south of the PFZ.

680
681 It is rewarding that the particle settling speeds obtained with the isopycnal mixing model
682 (which does not integrate the conservation equation over the water column) are in the range of
683 the speeds generally obtained with the vertical 1D model (see section 4.1; Rutgers van der
684 Loeff and Berger, 1993). In the 1D production-scavenging model, w_p is given by (see section
685 4.1.): $w_p = P/(d^{230}\text{Th}_p/dz)$. However, w_p is usually obtained by averaging $(d^{230}\text{Th}_p/dz)$ over
686 the whole water column or a large part of the water column as we did in section 4.1.

687 Consequently, the w_p does not depend on sharp changes of $(d^{230}\text{Th}_p/dz)$ that could result from
688 random errors on ²³⁰Th_p at intermediate depths. In the 1D mixing-scavenging model some
689 advection is introduced but the model is still integrated from the surface to the seafloor, so
690 that random errors on ²³⁰Th_p tend to be smoothed out (Rutgers van der Loeff and Berger,
691 1993). In addition, ²³⁴U *in situ* decay is not the only source of dissolved ²³⁰Th, because
692 advection also brings dissolved and particulate ²³⁰Th, which adds up uncertainty on the input
693 rate of ²³⁰Th.

694 In the isopycnal mixing model presented here, the uncertainties cited above can have much
695 larger effects on w_p because i) $(d^{230}\text{Th}_p/dz)$ is calculated over much thinner depth ranges and
696 ii) combined effects of high water flow rates and small ²³⁰Th concentration gradients implies
697 that the net effect of circulation on ²³⁰Th transport is subject to large relative uncertainties.
698 Nevertheless, the isopycnal mixing model opens the possibility to calculate the vertical

699 evolution of w_p in highly dynamical environments where the hypotheses of the 1D model are
700 clearly not fulfilled.

701

702 In a recent series of paper, non-linear dissolved ^{230}Th profiles are modelled with a
703 purely 1D approach by adjusting adsorption/desorption/remineralization rates and particle
704 settling velocities that are assumed to vary with depth (Lerner et al., 2016, 2017 and 2018).
705 These numerous parameters are adjusted by using different Th isotopes (^{228}Th , ^{230}Th , ^{234}Th).
706 This approach is different but not inconsistent with our approach because: (1) Unlike the
707 present paper, Lerner et al focus on areas where advection and eddy diffusion are expected to
708 be negligible; (2) In the present paper, we make no hypothesis on the processes driving
709 dissolved-particulate transfers (Eq. 7). Ultimately, it should be possible to determine
710 adsorption/desorption rates in areas with weak currents and then to use these values in regions
711 with a vigorous circulation such as the ACC. Lerner's approach treats explicitly the case of w_p
712 varying with depth, whereas our approach to leave the possibility to determine the particle
713 settling speed at different depths/density. To do so rigorously, it will be necessary to treat w_p
714 like a parameter varying with depth (see section 4.3).

715

716 5. Conclusion

717 While we still used crude approximations, the model proposed here opens the
718 possibility to determine the particle settling speed and to bring constraints on the isopycnal
719 mixing rate at different vertical levels and with a more realistic description of the vertical
720 structure of the water column. The improvement that will come out of the method proposed
721 here will require sampling with higher horizontal and vertical sampling resolutions, a goal
722 partly achieved in the framework of the GEOTRACES program. It also requires progresses
723 for the particulate Th analysis and constant efforts for the intercalibration of the different
724 cruises.

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918 Table ES1: Dissolved Thorium data.

Station	Depth (m)	θ (°C)	Salinity	γ^n (kg/m ³)	²³² Th _d ¹ (pg/kg)	²³⁰ Th _p ¹ (pg/kg)	²³⁰ Th _{p-xs} ² (pg/kg)	Water mass
S1: 36.50°S, 13.10°E, 4923 m bottom depth								
	29	20.785	35.617	25.0222	34.7 ± 1.3	1.21 ± 0.03	1.06 ± 0.03	ICW
	198	12.364	35.014	26.53	52.1 ± 1.3	2.48 ± 0.04	2.25 ± 0.04	ICW
	397	9.824	34.757	26.7918	50.1 ± 1.5	4.52 ± 0.06	4.30 ± 0.06	ICW
	753	4.973	34.346	27.1602	37.2 ± 1.3	5.45 ± 0.08	5.29 ± 0.08	i-AAIW
	1188	3.279	34.551	27.5004	38.0 ± 1.3	7.38 ± 0.12	7.22 ± 0.12	i-AAIW
	1975	2.515	34.808	27.7752	40.2 ± 1.4	11.64 ± 0.18	11.46 ± 0.18	SE-NADW
	2664	2.132	34.838	27.8316	42.0 ± 1.3	14.71 ± 0.19	14.52 ± 0.19	SE-NADW
	3005	1.945	34.836	27.8444	42.2 ± 1.4	14.94 ± 0.17	14.76 ± 0.17	SE-NADW
	3981	0.892	34.749	27.8506	70.0 ± 1.4	17.55 ± 0.23	17.25 ± 0.23	AABW
	4565	0.716	34.733	27.8493	69.1 ± 1.6	21.70 ± 0.27	21.40 ± 0.27	AABW
	4907	0.568	34.72	27.8481	288.5 ± 3.5	37.10 ± 0.41	35.84 ± 0.41	AABW
S2: 42.47°S, 08.93°E, 4070 m bottom depth								
	20	12.951	34.521	26.0547	11.1 ± 1.4	0.74 ± 0.02	0.70 ± 0.03	ICW
	124	9.083	34.482	26.7815	19.8 ± 1.4	1.84 ± 0.03	1.75 ± 0.04	ICW
	292	6.654	34.267	27.0053	36.2 ± 1.3	3.04 ± 0.04	2.89 ± 0.04	ICW/a-AAIW
	398	6.01	34.292	27.1231	38.8 ± 1.5	4.13 ± 0.06	3.97 ± 0.06	ICW/a-AAIW
	595	4.138	34.185	27.2829	30.9 ± 1.4	5.14 ± 0.08	5.01 ± 0.08	a-AAIW
	940	3.075	34.313	27.5162	25.7 ± 1.3	7.02 ± 0.09	6.90 ± 0.09	a-AAIW
	1453	2.626	34.585	27.7945	26.1 ± 1.3	9.19 ± 0.11	9.07 ± 0.11	UCDW
	2172	2.346	34.79	27.994	31.1 ± 1.3	13.36 ± 0.16	13.22 ± 0.16	SE-NADW
	2905	1.967	34.827	28.0789	35.8 ± 1.3	14.43 ± 0.16	14.28 ± 0.16	SE-NADW
	3445	1.427	34.792	28.1359	36.9 ± 1.6	16.13 ± 0.24	15.96 ± 0.24	SE-NADW
	4021	0.81	34.741	28.1961	63.9 ± 1.5	17.96 ± 0.22	17.68 ± 0.22	AABW
S3: 47.55°S, 04.37°E, 4480 m bottom depth								
	40	6.32	33.733	26.5685	7.5 ± 1.4	1.19 ± 0.04	1.15 ± 0.04	AASW
	124	4.029	33.867	27.0072	8.3 ± 1.3	1.82 ± 0.04	1.79 ± 0.04	AASW
	248	3.633	34.101	27.2697	15.3 ± 1.4	3.55 ± 0.07	3.48 ± 0.07	AASW/a-AAIW
	495	2.739	34.233	27.4888	17.1 ± 1.2	5.07 ± 0.07	5.00 ± 0.07	a-AAIW
	742	2.591	34.407	27.6574	16.8 ± 1.3	6.46 ± 0.11	6.39 ± 0.11	a-AAIW/UCDW
	1068	2.447	34.575	27.8099	18.2 ± 1.3	8.26 ± 0.10	8.18 ± 0.10	A_UCDW
	1482	2.334	34.716	27.936	22.2 ± 1.5	10.93 ± 0.17	10.83 ± 0.17	SW-NADW
	2003	1.971	34.771	28.0376	23.2 ± 1.2	12.68 ± 0.15	12.58 ± 0.15	SW-NADW
	2003				23.2 ± 1.5	13.45 ± 0.16	13.34 ± 0.16	
	3052	0.955	34.731	28.1653	29.2 ± 1.6	14.94 ± 0.19	14.81 ± 0.19	SW-NADW
	4299	0.415	34.695	28.2262	18.5 ± 1.3	8.83 ± 0.12	8.75 ± 0.12	AABW

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S4: 51.85°S, 00.00°E, 2570 m bottom depth								
	59	2.521	33.712	27.0277	4.7 ± 1.5	1.20 ± 0.03	1.18 ± 0.03	AASW
	168	0.827	34.033	27.4838	10.8 ± 1.5	2.82 ± 0.06	2.77 ± 0.06	WW (AASW)
	248	1.543	34.433	27.7832	13.4 ± 1.6	5.04 ± 0.08	4.98 ± 0.08	WW/DP-UCDW
	327	1.811	34.548	27.8553	13.8 ± 1.6	6.64 ± 0.10	6.58 ± 0.10	DP-UCDW
	416	1.874	34.627	27.9165	17.4 ± 1.3	8.68 ± 0.10	8.61 ± 0.10	DP-UCDW
	416	1.874	34.627	27.9165	15.7 ± 1.3	7.94 ± 0.10	7.87 ± 0.10	DP-UCDW
	742	1.702	34.704	28.012	21.5 ± 1.5	12.16 ± 0.14	12.06 ± 0.14	LCDW
	1117	1.42	34.727	28.08	27.3 ± 1.3	15.28 ± 0.17	15.16 ± 0.17	LCDW
	1678	0.755	34.704	28.171	28.6 ± 1.5	16.70 ± 0.19	16.58 ± 0.19	LCDW/AABW
	2307	0.359	34.689	28.225	30.5 ± 1.4	18.11 ± 0.21	17.98 ± 0.21	AABW
	2501	0.319	34.687	28.231	30.1 ± 1.3	16.82 ± 0.19	16.69 ± 0.19	AABW
Intercal: 52.98°S, 00.00°W, 2624 m bottom depth								
	380	1.81	34.63	27.929	19.1 ± 1.2	9.93 ± 0.13	9.85 ± 0.13	UCDW
	380	1.81	34.63	27.929	19.6 ± 1.4	9.90 ± 0.12	9.82 ± 0.12	UCDW
	500	1.78	34.68	27.979	22.0 ± 1.2	12.14 ± 0.14	12.05 ± 0.14	LCDW
	500	1.78	34.68	27.979	24.5 ± 1.5	13.44 ± 0.20	13.33 ± 0.20	LCDW
	1000	1.22	34.71	28.094	27.4 ± 1.2	16.08 ± 0.20	15.96 ± 0.20	LCDW
	1000	1.22	34.71	28.094	27.5 ± 1.3	16.11 ± 0.18	15.99 ± 0.18	LCDW
S5: 57.55°S, 00.03°W, 3932 m bottom depth								
	134	-0.671	34.355	27.8713	19.7 ± 1.5	8.26 ± 0.11	8.17 ± 0.11	WW (AASW)
	247	0.353	34.63	28.094	29.4 ± 1.5	15.64 ± 0.19	15.51 ± 0.19	WW (AASW)/ WDW
	396	0.529	34.674	28.154	32.8 ± 1.4	19.10 ± 0.24	18.96 ± 0.24	WDW
	692	0.422	34.685	28.202	36.1 ± 1.5	23.00 ± 0.24	22.84 ± 0.24	WDW
	1185	0.154	34.677	28.2434	42.2 ± 1.5	26.59 ± 0.29	26.41 ± 0.29	WDW / WSDW
	1776	-0.104	34.668	28.2872	49.4 ± 1.6	30.31 ± 0.32	30.10 ± 0.32	WDW / WSDW
	2462	-0.346	34.66	28.332	60.6 ± 1.4	34.30 ± 0.36	34.04 ± 0.36	WSDW
	3147	-0.502	34.655	28.3619	71.5 ± 1.6	34.42 ± 0.36	34.10 ± 0.36	WSDW
	3848	-0.632	34.65	27.8713	83.1 ± 1.6	33.93 ± 0.37	33.57 ± 0.37	WSDW / WSBW
	3848	-0.632	34.65	27.8713	82.5 ± 1.8	33.78 ± 0.38	33.42 ± 0.38	WSDW / WSBW

928 ¹: Uncertainties are 2σ_n (error on the mean). ²: The excess ²³⁰Th_{d-xs} is calculated using the
 929 equation:

$$930 \quad {}^{230}\text{Th}_{d-xs} = {}^{230}\text{Th}_{d-measured} - {}^{232}\text{Th}_{d-measured} \times \left(\frac{{}^{230}\text{Th}}{{}^{232}\text{Th}} \right)_{\text{Lithogenic}}, \text{ where the}$$

931 $({}^{230}\text{Th}/{}^{232}\text{Th})_{\text{Lithogenic}} = 4.0 \times 10^{-6}$ (mol/mol), close to average crust ${}^{230}\text{Th}/{}^{232}\text{Th} = 4.1 \times 10^{-6}$.

932 The lithogenic correction is small: generally less than 2% of ²³⁰Th_d and occasionally reaching
 933 6 to 11% in the surface waters of S2 and S1 respectively. Therefore the ²³⁰Th_d and ²³⁰Th_{d-xs}
 934 profiles do not differ significantly.

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Table ES2: Particulate thorium data (in situ pump filtration)

Station	Depth (m)	Volume ¹ (L)	²³² Th _p (pg/kg) ²	²³⁰ Th _p (pg/kg) ²	²³⁰ Th _{p-xs} (pg/kg) ²	²³⁰ Th _{p-xs} particulate fraction
S1	30	346	7.0 ± 0.1	0.076 ± 0.002	0.045 ± 0.003	4 %
	199	803	18.8 ± 0.3	0.209 ± 0.004	0.128 ± 0.004	6 %
	1242	997	32.3 ± 0.7	1.45 ± 0.03	1.31 ± 0.03	18 %
	2687	142	37.8 ± 0.5	1.309 ± 0.003	1.144 ± 0.004	8 %
	2732	1202	17.3 ± 0.5	1.8 ± 0.4	1.7 ± 0.4	12 %
	4628	610	371 ± 10	28.1 ± 0.4	26.5 ± 0.4	124 %
	S2	20	315	4.1 ± 0.1	0.148 ± 0.004	0.130 ± 0.004
257		814	8.9 ± 0.1	0.379 ± 0.003	0.340 ± 0.003	12 %
601		961	5.4 ± 0.1	0.492 ± 0.004	0.469 ± 0.004	9 %
1426		1053	9.1 ± 0.1	1.462 ± 0.009	1.422 ± 0.009	16 %
1941		531	8.5 ± 0.1	0.752 ± 0.005	0.715 ± 0.005	5 %
2859		515	8.9 ± 0.1	1.63 ± 0.01	1.59 ± 0.01	11 %
3858		508	93.1 ± 1.2	8.51 ± 0.07	8.10 ± 0.07	48 %
S3		40	99	7.1 ± 0.1	0.163 ± 0.002	0.133 ± 0.002
	594	500	4.41 ± 0.04	0.686 ± 0.002	0.667 ± 0.002	13 %
	1058	967	5.1 ± 0.1	1.045 ± 0.003	1.023 ± 0.003	12 %
	1985	1128	5.5 ± 0.1	1.426 ± 0.004	1.402 ± 0.004	13 %
	3025	218	7.7 ± 0.1	1.778 ± 0.006	1.745 ± 0.006	12 %
	3024	552	8.0 ± 0.1	1.838 ± 0.005	1.804 ± 0.005	12 %
	4262	210	2.5 ± 0.0	0.344 ± 0.002	0.333 ± 0.002	4 %
	S4	59	381	0.076 ± 0.001	0.0202 ± 0.0005	0.0198 ± 0.005
169		702	1.48 ± 0.02	0.300 ± 0.001	0.294 ± 0.001	11 %
258		903	2.42 ± 0.02	0.467 ± 0.001	0.457 ± 0.001	9 %
743		984	2.23 ± 0.02	0.682 ± 0.002	0.672 ± 0.002	6 %
1119		1069	2.82 ± 0.03	0.630 ± 0.002	0.617 ± 0.002	4 %
1120		308	1.72 ± 0.02	0.368 ± 0.002	0.360 ± 0.002	2 %
1682		1018	2.23 ± 0.02	0.727 ± 0.002	0.717 ± 0.002	4 %
2273		938	6.07 ± 0.06	1.885 ± 0.005	1.858 ± 0.005	10 %
2469		493	6.25 ± 0.06	1.850 ± 0.005	1.823 ± 0.005	11 %
S5		30	341	2.23 ± 0.01	0.706 ± 0.005	0.697 ± 0.005
	139	872	2.1 ± 0.1	0.261 ± 0.002	0.251 ± 0.002	3 %
	248	927	2.5 ± 0.1	0.507 ± 0.003	0.496 ± 0.003	3 %
	694	127	3.6 ± 0.8	0.555 ± 0.009	0.539 ± 0.010	2 %
	1784	290	4.0 ± 0.4	0.714 ± 0.006	0.697 ± 0.006	2 %
	2478	1066	8.5 ± 0.1	1.726 ± 0.029	1.69 ± 0.03	5 %
	3172	905	11.6 ± 0.2	1.754 ± 0.006	1.703 ± 0.006	5 %
	3840	715	26.1 ± 0.3	2.966 ± 0.012	2.852 ± 0.012	8 %

940 ¹ Fraction of the volume of filtered water used for Thorium analysis. ² Uncertainties are $2\sigma_n$
941 (error on the mean).
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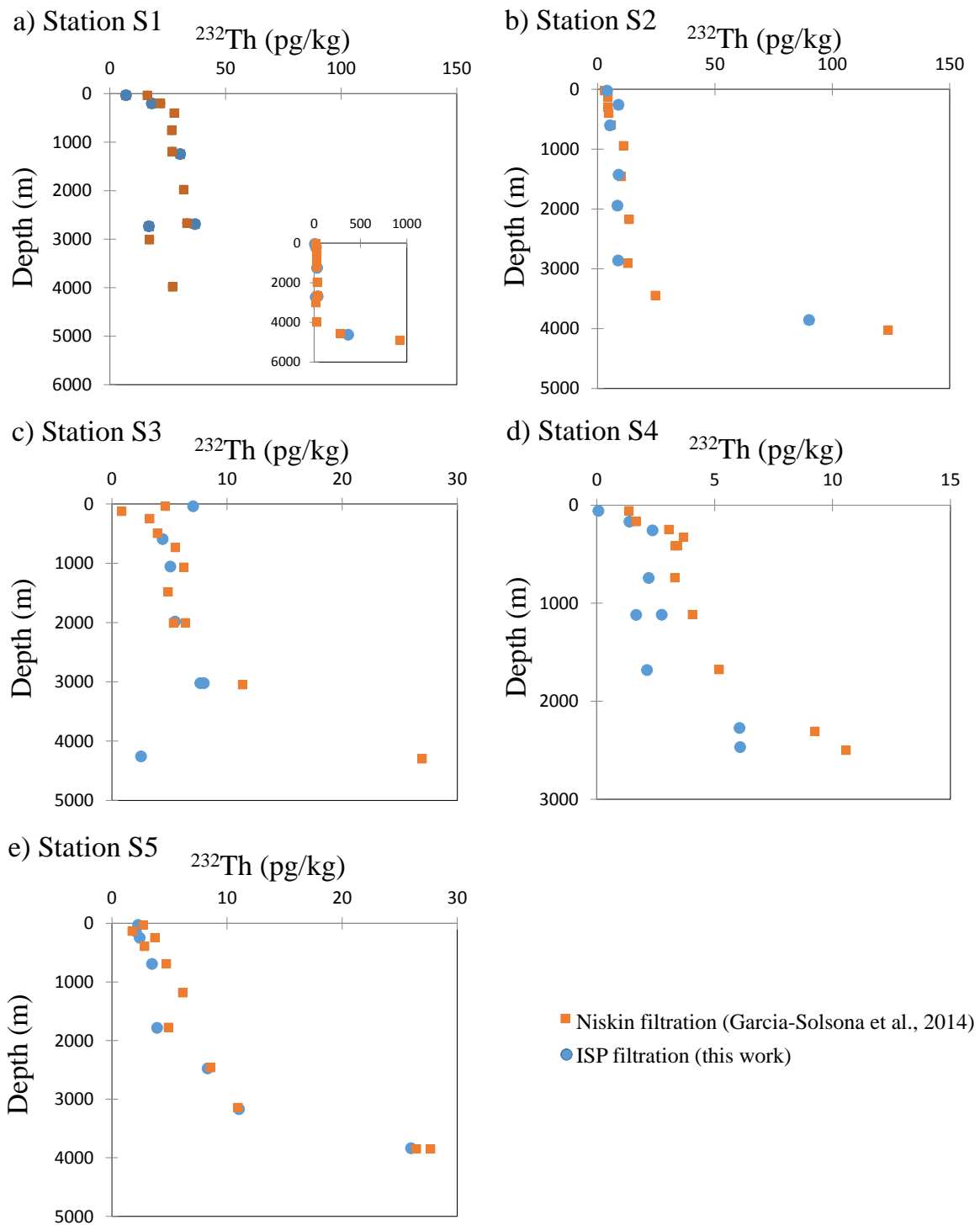
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Table ES3: Total thorium isotopes recalculated on neutral density surfaces

	Depth (m)	θ (°C)	Salinity	^{232}Th (pg/kg)	error	^{230}Th (fg/kg)	error
$\gamma^n = 27.865 \text{ kg/m}^3$							
S1	-1550	2.732	34.695	10.67	0.15	72.4	1.5
S2	-1631	2.564	34.667	11.51	0.12	36.3	1.3
S3	-1227	2.44	34.644	10.07	0.13	25.1	2.9
S4	-345	1.838	34.563	7.59	0.11	16.8	1.8
S5	-98	-0.665	34.348	8.52	0.11	21.8	0.1
$\gamma^n = 28.094 \text{ kg/m}^3$							
S1	3073	1.86	34.827	15.1	0.2	60.7	1.5
S2	3082	1.849	34.823	17.3	0.8	52.9	0.2
S3	2440	1.722	34.756	15.2	0.2	32.1	1.3
S4	1205	1.322	34.726	15.9	0.2	29.3	1.3
intercal	-999.6	1.205	34.71	16.7	0.2	30.1	1.2
intercal	-999.6	1.205	34.71	16.7	0.2	30.2	1.3
S5	247	0.28616	34.619	16.1	0.2	31.9	1.6

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947 Fig. ES1: comparison of particulate ^{232}Th collected by in situ pump and Niskin filtrations.

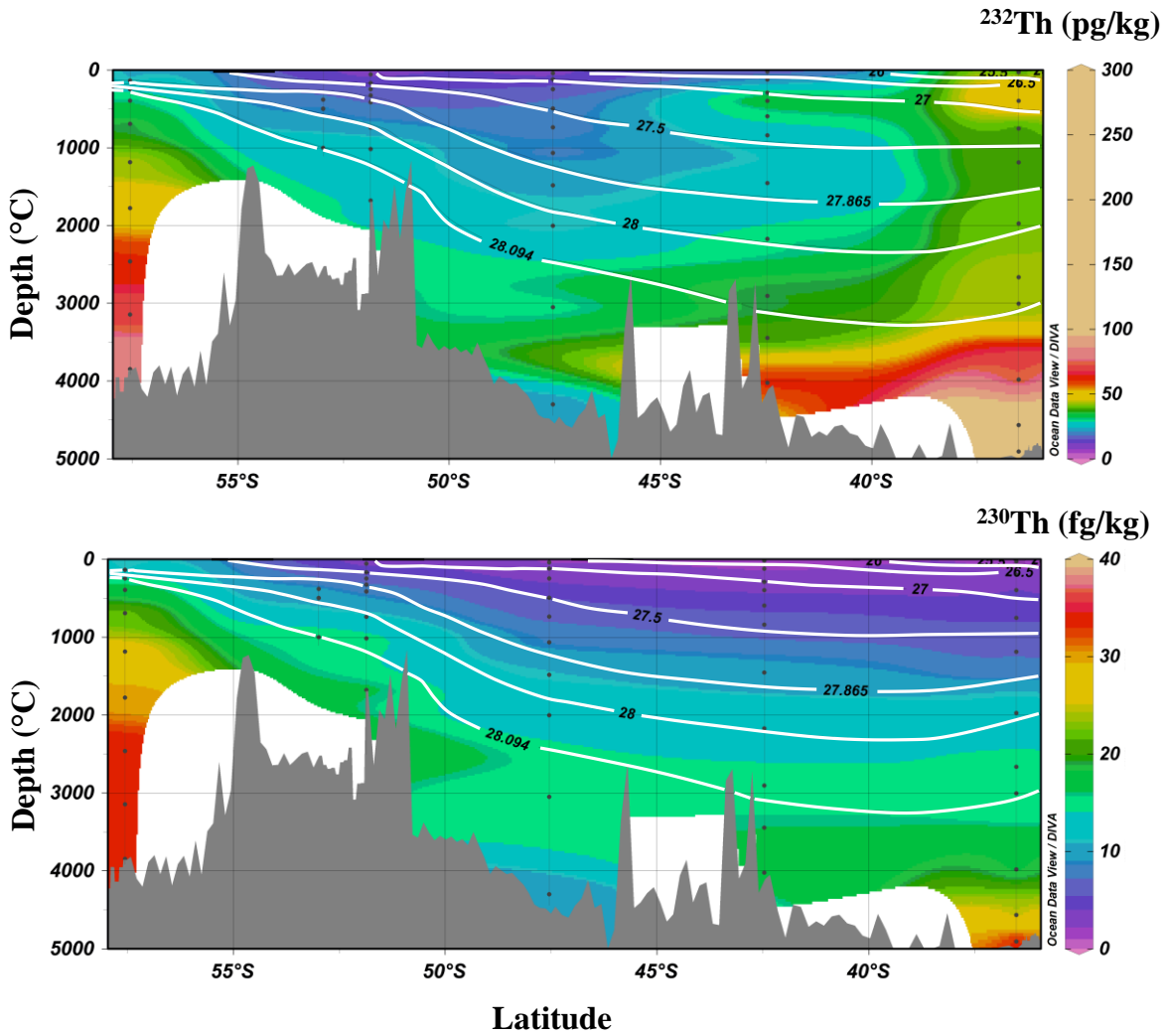


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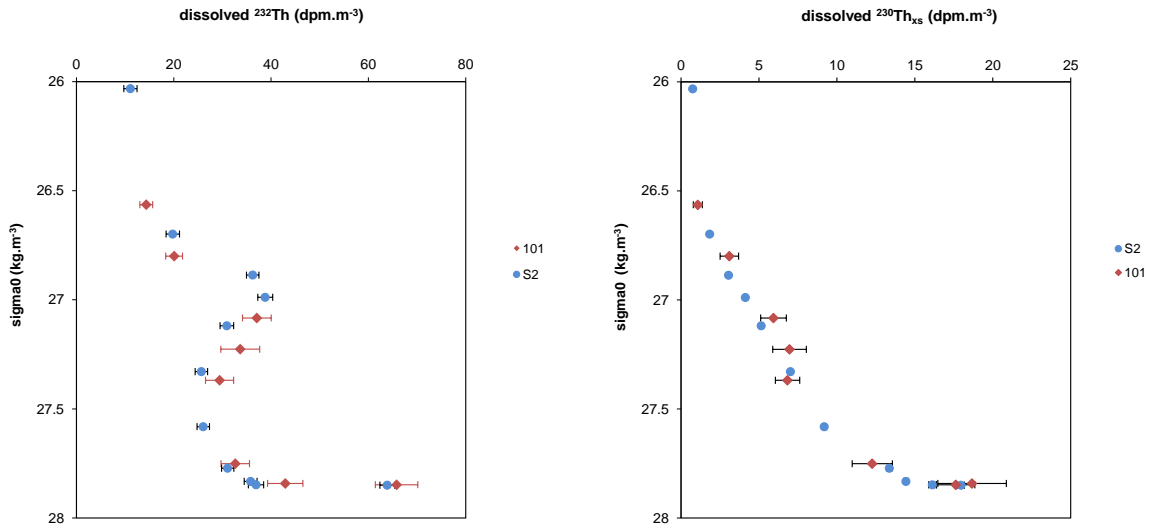
Figure ES2: Dissolved Th isotopes BGH sections. (a) $^{232}\text{Th}_d$. (b): $^{230}\text{Th}_d$. White curves: neutral density surfaces.



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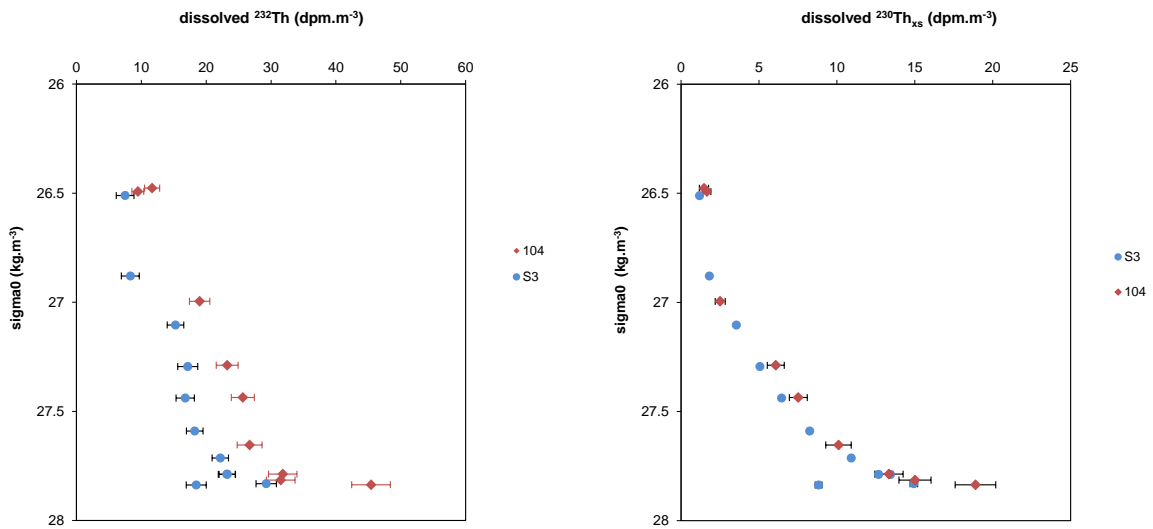
964 Figure ES3: Dissolved Th comparison between 2 stations of the BGH and Zero-Drake
965 sections. $^{232}\text{Th}_d$ and $^{230}\text{Th}_d$ are plotted versus potential density anomaly in order to reduce
966 hydrographical variations between the 2 cruises.
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