

1 **Meridional circulation across the Antarctic Circumpolar Current serves as a**
2 **double ^{231}Pa and ^{230}Th trap**

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19 **Abstract**

20 Upwelling of Circumpolar Deep Water in the Weddell Gyre and low scavenging rates south of the

21 Antarctic Circumpolar Current (ACC) cause an accumulation of particle reactive nuclides in the Weddell

22 Gyre. A ventilation/reversible scavenging model that successfully described the accumulation of ^{230}Th in

23 this area was tested with other particle reactive nuclides and failed to adequately describe the depth-

24 distributions of ^{231}Pa and ^{210}Pb . We present here a modified model that includes a nutrient-like

25 accumulation south of the Antarctic Polar Front in an upper meridional circulation cell, as well as

26 transport to a deep circulation cell in the Weddell Gyre by scavenging and subsequent release at depth.

27 The model also explains depletion of ^{231}Pa and ^{230}Th in Weddell Sea Bottom Water (WSBW) by
28 ventilation of newly formed deep water on a timescale of 10 years, but this water mass is too dense to
29 leave the Weddell Gyre.

30 In order to quantify the processes responsible for the ^{231}Pa - and ^{230}Th - composition of newly formed
31 Antarctic Bottom Water (AABW) we present a mass balance of ^{231}Pa and ^{230}Th in the Atlantic sector of
32 the Southern Ocean based on new data from the GEOTRACES program. The ACC receives $6.0 \pm 1.5 \times 10^6$
33 dpm s^{-1} of ^{230}Th from the Weddell Sea, similar in magnitude to the net input of $4.2 \pm 3.0 \times 10^6 \text{ dpm s}^{-1}$
34 from the north. For ^{231}Pa , the relative contribution from the Weddell Sea is much smaller, only $0.3 \pm 0.1 \times$
35 10^6 , compared to $2.7 \pm 1.4 \times 10^6 \text{ dpm s}^{-1}$ from the north. Weddell Sea Deep Water (WSDW) leaving the
36 Weddell Gyre northward to form AABW is exposed in the ACC to resuspended opal-rich sediments that
37 act as efficient scavengers with a Th/Pa fractionation factor $F \leq 1$. Hydrothermal inputs may provide
38 additional removal with low F. Scavenging in the full meridional circulation across the opal-rich ACC thus
39 acts as a double ^{231}Pa and ^{230}Th trap that preconditions newly formed AABW.

40

41 **Keywords**

42 ^{231}Pa

43 ^{230}Th

44 Weddell Sea

45 Antarctic Bottom Water

46 GEOTRACES

47 scavenging

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52 Introduction

53 The pair of long-lived radionuclides ^{231}Pa and ^{230}Th is well suited for paleoceanographic interpretations.
54 These nuclides are produced at a constant rate in the water from their respective U parents, ^{235}U and
55 ^{234}U . Both are rapidly removed from the water column by scavenging but differences in their reactivities
56 cause the two to be fractionated by particle flux, particle composition, and ocean circulation with the
57 result that their activity ratio in surface sediments deviates from the production ratio of 0.093
58 (Anderson et al., 1983). The original interpretation of ^{231}Pa and ^{230}Th accumulation in sediments of the
59 Southern Ocean was based on boundary scavenging. DeMaster (1981) interpreted high ^{231}Pa and ^{230}Th
60 accumulation rates in the opal belt of the South Atlantic as indicative of high productivity, and the
61 $^{231}\text{Pa}/^{230}\text{Th}$ ratio in Holocene and LGM sediments was used by Kumar et al. (1995) as indication for
62 glacial-interglacial changes in the zones of high productivity. Yu et al. (1996) demonstrated that ^{231}Pa
63 produced in the Atlantic Ocean was deposited in the opaline sediments of the ACC, paving the way to
64 use the isotope ratio as a proxy for meridional overturning circulation (MOC).

65

66 This proxy builds on the idea that the $^{231}\text{Pa}/^{230}\text{Th}$ ratio in deep waters increases with water mass age and
67 that it is reflected in the $^{231}\text{Pa}/^{230}\text{Th}$ ratio in deposited sediments. Glacial-Interglacial changes in
68 $^{231}\text{Pa}/^{230}\text{Th}$ ratios in North Atlantic sediments have been interpreted to show changes in North Atlantic
69 Deep Water (NADW) flow (McManus et al., 2004). Cores recovered from the South Atlantic at 2440 and
70 3213m, now bathed in NADW, were covered by Southern Component Water (SCW) during the Last
71 Glacial Period (LGP) and their LGP $^{231}\text{Pa}/^{230}\text{Th}$ ratios have been interpreted in terms of northward flow
72 (Jonkers et al., 2015; Negre et al., 2010). Similarly, south Atlantic cores from greater depth are expected
73 to record the history of AABW flow (Lippold et al., 2016). Interpretations in terms of northward flow of
74 SCW or AABW depend on assumptions on the scavenging history and resulting preformed $^{231}\text{Pa}/^{230}\text{Th}$

75 ratio in these southern source waters (Jonkers et al., 2015; Lippold et al., 2016; Negre et al., 2010).
76 Considering that the $^{231}\text{Pa}/^{230}\text{Th}$ ratio in newly deposited particles is largely determined by the lower
77 1000m of the water column (Thomas et al., 2006) and that wide areas of the abyssal sediments are
78 bathed in AABW, it is important to know what controls the ^{231}Pa and ^{230}Th activities in freshly produced
79 AABW. A large part of AABW enters the ACC from the Weddell Gyre as Weddell Sea Deep Water
80 (WSDW), whether it is produced in the Gyre (according to a traditional estimate 60-70% of AABW, Orsi
81 et al., 1999; Orsi et al., 2002) or advected in the coastal current from the Indian Ocean (Hoppema et al.,
82 2001; Jullion et al., 2014; Meredith et al., 2000). The processes controlling the $^{231}\text{Pa}/^{230}\text{Th}$ ratio in the
83 WSDW are therefore important for the interpretation of the ratio in deep water sediments at lower
84 latitudes as proxy for MOC (Anderson et al., submitted in parallel).

85 According to the classical reversible scavenging model, the activities of dissolved and particulate $^{230}\text{Th}_{\text{xs}}$
86 and $^{231}\text{Pa}_{\text{xs}}$ increase linearly with depth (Bacon and Anderson, 1982; Nozaki et al., 1981), where the
87 subscript xs means excess activities, i.e. the activities of nuclides produced by U decay in seawater.
88 Although all activities presented here are excess activities, we will, following Deng et al. (2014), leave
89 out the subscript in this paper. Rutgers van der Loeff and Berger (1993) reported ^{230}Th and ^{231}Pa profiles
90 from the Weddell Gyre that reached maximum concentrations at mid-depth, and showed how the
91 distribution of these isotopes was controlled by upwelling of CDW and low scavenging rates in these
92 waters south of the ACC characterized by low particle flux (Fischer et al., 1988).

93 The scavenging of Pa depends strongly on the opal flux. The F ratio (scavenging preference of Th over
94 Pa) changes dramatically through the ACC. North of the ACC the F ratio is 10 or higher, whereas in the
95 opal belt and south of it Pa is much more strongly scavenged than further north, giving an F ratio <1
96 (Chase et al., 2002; Walter et al., 1997). The change in F ratio across the ACC is a circumpolar
97 phenomenon. Chase et al. (2003) gave a detailed description of the change in the south Pacific based on

98 water, surface sediment and sediment trap data. They showed that, in this area of the Pacific with little
99 deep water formation, the activities of ^{230}Th and ^{231}Pa show no significant gradients on isopycnal
100 surfaces implying that the upwelling and isopycnal ventilation are too rapid to allow changes in nuclide
101 activities in the upwelled water masses. In the Atlantic sector, upwelled deep waters have a long
102 residence time in the Weddell Gyre under a low-scavenging regime, and although we know that ^{230}Th
103 accumulates under these circumstances (Rutgers van der Loeff and Berger, 1993), it has not yet been
104 clearly shown whether the low scavenging regime with a low F ratio also allows ^{231}Pa to accumulate.
105 Recently, new data on the distribution of ^{230}Th , ^{231}Pa and ^{232}Th have become available in the GEOTRACES
106 program. Venchiarutti et al. (2011a) described the distribution of these nuclides in Drake Passage and so
107 constrained the composition of Pacific waters transported in the ACC. Deng et al. (2014) quantified the
108 transport of ^{231}Pa and ^{230}Th with NADW into the Southern Ocean. Venchiarutti et al. (2011b) gave new
109 data from Weddell Gyre and Zero Meridian. With this widely improved dataset we now have a firm basis
110 to address the following questions:

- 111 - What controls the accumulation of ^{230}Th and ^{231}Pa in the Weddell Gyre and especially in WSDW?
- 112 - What controls the composition of ^{230}Th and ^{231}Pa in AABW exported from the Weddell Gyre and
113 across the ACC to the abyssal ocean?
- 114 - What is the fate of the ^{230}Th and ^{231}Pa imported in the SO with NADW (Deng et al., 2014) and
115 what is the mass balance of ^{230}Th and ^{231}Pa in the Atlantic sector of the Southern Ocean?

116 In this paper, we present a model to explain the observed distributions of ^{230}Th and ^{231}Pa in the Weddell
117 Gyre, check the model by applying it to another particle reactive nuclide (^{210}Pb) and give a mass balance
118 of ^{230}Th and ^{231}Pa in the Atlantic sector of the Southern Ocean. We argue that the ACC functions as a
119 double trap for ^{230}Th and ^{231}Pa , once in the surface water and once again in the outflowing deep water.

120

121 **Hydrography of the Weddell Gyre and adjacent ACC:**

122 In the Southern Ocean, shoaling Circumpolar Deep Water (CDW) feeds into a shallow meridional
123 overturning cell, upwelling into surface waters and leading to the formation of intermediate waters, and
124 into a deep cell transforming CDW and forming Antarctic Bottom Water (Chase et al., 2003; Fahrbach et
125 al., 2011, Fig. 2). The transport is overwhelmed by the zonal transport in the ACC.

126 In the Weddell Gyre, CDW enters from the east and circulates as what is locally known as Warm Deep
127 Water (WDW) at depths of 200-1500m. WDW with at its core a neutral density anomaly of 27.88 kg m^{-3}
128 is primarily of Lower Circumpolar Deep Water (LCDW) density. Dense waters are produced by ice
129 formation on the continental shelf (high salinity shelf water, HSSW) and under the ice shelf (Ice Shelf
130 Water, ISW). WDW mixes with these salinified and cooled shelf waters to form Weddell Sea Bottom
131 Water (WSBW) and the somewhat less dense WSDW. When these newly formed deep waters leave the
132 Weddell Gyre they are the source of the circumpolar AABW that fills the abyssal ocean.

133 The Weddell-Enderby basin has a cyclonic circulation with a strong recirculation in the western part
134 (Fahrbach et al., 2011). The basin is confined by a ridge system in the north, and deep and bottom
135 waters can only leave the basin through gaps in these ridges. Outflows of WSDW to the ACC and
136 beneath the ACC to the Argentine Basin are observed over the south Scotia Sea and Georgia Basin, and
137 over the Enderby Basin toward the Crozet-Kerguelen Gap (Haine et al., 1998; Orsi et al., 1999; Orsi et al.,
138 1993) (Fig. 1). About half of the export of WSDW occurs through the gaps in the Scotia Ridge (Jullion et
139 al., 2014; Naveira Garabato et al., 2002). This deep water, that exits through the Georgia Passage and
140 Georgia Basin into the Argentine Basin, is more ventilated, cooler and fresher, with origins rather on the
141 slope of the Antarctic Peninsula, whereas the WSDW leaving further east is derived from the Filchner-
142 Ronne Ice Shelf (Gordon et al., 2001; Naveira Garabato et al., 2002).

143

144 **Upwelling, circulation and scavenging: the behavior of particle-reactive nuclides in the Weddell Gyre**

145 The distribution of ^{230}Th in the Weddell Gyre has been measured during Polarstern expedition ANT-VIII/3
146 in 1991 by Rutgers van der Loeff and Berger (1993) and more recently during Polarstern expedition ANT-
147 XXIV/3 (PS71) in 2008 by (Venchiarutti et al., 2011a; Venchiarutti et al., 2011b) (Fig 1). Positions and
148 references for all stations discussed in this paper are given in Table S1. The ^{230}Th distribution in the
149 Weddell Gyre was explained by Rutgers van der Loeff and Berger (1993) using a model that combined a
150 simple representation of southward upwelling of ^{230}Th -rich LCDW across the ACC in the Antarctic
151 divergence with reduced scavenging in relation to the very low particle fluxes in the Weddell Gyre.
152 Further evidence for this reduced scavenging rate was provided by sediment trap data (Fischer et al.,
153 1988) and sediment trap and - core analyses by Walter et al. (2000).

154 In addition, it was shown that in the deepest water layer where potential temperature $\theta < -0.7^\circ\text{C}$ and the
155 water mass is characterized by Weddell Sea Bottom Water, ^{230}Th activities are lower than at mid-depth
156 (observed both in the 1991 and the 2008 dataset), a feature not represented in the model of Rutgers
157 van der Loeff and Berger (1993). In principle, this decline can be caused by two mechanisms: either by
158 seafloor scavenging, as has been proposed in several other deep-sea environments (Deng et al., 2014;
159 Hayes et al., 2015a; Jeandel et al., 2015), or by ventilation with new bottom water formation as already
160 suggested by Rutgers van der Loeff and Berger (1993). Fig. 3 shows the effect of ventilation by bottom
161 water formation on the distribution of ^{230}Th compared with ^{230}Th data of station 193 of Polarstern
162 expedition ANTXXIV-3 in 2008 (Venchiarutti et al., 2011b) (Fig.1, Table S1). The composition of the
163 bottom water was derived from an Optimum Multiparameter Analysis (OMP) based on potential
164 temperature, salinity, helium isotope, neon data collected during ANTXXIV-3 (applying the same
165 approach and parameter setting to the 2008 data as described in Huhn et al., 2008). This analysis
166 showed that in the vicinity of station 193, at station 196 (datapoint with available He isotope and Ne

167 data closest to station 193, Fig. 1, Table S1) the bottom water consisted of $46\pm 5\%$ Warm Deep Water
168 (WDW), $38\pm 13\%$ High Salinity Shelf Water (HSSW), $8\pm 7\%$ Ice Shelf Water (ISW) and $8\pm 3\%$ surface water.
169 For WDW with potential temperature of 0.44°C , we interpolate from the experimental profile a
170 dissolved ^{230}Th concentration of 1.14 dpm m^{-3} . For all surface water components, including ISW and
171 HSSW, we use the ^{230}Th activity that we found at 50m depth on Sta 193 (0.18 dpm m^{-3}), which compares
172 well with the activities found by Fleisher and Anderson (2003) at the end of summer in the Ross Sea. For
173 recently ventilated WSBW, we find a dissolved ^{230}Th activity of 0.65 dpm m^{-3} . In Fig.3, we show that it
174 takes about 10 years before ingrowth and reversible exchange with settling particles cause this activity
175 to increase to the measured value of 1.2 dpm m^{-3} . This result is in reasonable agreement with the
176 transient tracer (chlorofluorocarbons, CFC) based Transit Time Distribution as calculated by Huhn et al.
177 (2013), which gives the most likely age of 20 ± 5 years of the bottom water at station 193.

178 Now that the ^{230}Th data can be reasonably well explained in this way (Fig.3), we check whether the
179 model also adequately describes the distributions of other particle-reactive nuclides. We therefore
180 apply the model to ^{231}Pa and ^{210}Pb , using upwelling of deep waters north of the Antarctic Polar Front
181 (APF) with the composition as given by Venchiarutti et al. (2011b) for ^{231}Pa and by Somayajulu and Craig
182 (1976) for ^{226}Ra and ^{210}Pb . We use a vertical mixing rate of $5 \times 10^3\text{ m}^2\text{ y}^{-1}$ and for ^{210}Pb an atmospheric
183 input of $3\text{ dpm cm}^{-2}\text{ y}^{-1}$ (Rutgers van der Loeff and Berger, 1991), but for simplicity, the input of WSBW is
184 not included in these model runs shown in Fig. 4 (black lines). The scavenging rates of Pa and Pb are
185 based on that of Th using for Pa a fractionation factor of $F(\text{Th}/\text{Pa})=1$ (Walter et al., 1997) and for Pb in
186 the test run $F(\text{Th}/\text{Pb})=1$.

187 As shown from the model runs in Fig.4 (black lines), the fits for ^{231}Pa and ^{210}Pb to the experimental
188 values found in the Weddell Sea for ^{231}Pa (Venchiarutti et al., 2011b, Sta 193) and for ^{210}Pb (Somayajulu
189 and Craig, 1976, GEOSECS Station 89) are very poor. Indeed, the model developed for ^{230}Th fails here to

190 reproduce the very shallow maximum in dissolved ^{210}Pb and produces far too low activities of ^{231}Pa . In
191 order to improve the model and especially to better represent the much shallower accumulation of ^{231}Pa
192 and ^{210}Pb compared to ^{230}Th we must investigate in more detail what controls the scavenging process in
193 the Weddell Gyre.

194 **A closer look at particle rain and mineralization in the Weddell Gyre:**

195 *shallow mineralization*

196 The sedimentary record shows high recent opal sedimentation rates just south of the Southern Polar
197 Front (Geibert et al., 2005) and it has generally been assumed that productivity and export fluxes are
198 higher in this area than further south in the Weddell Sea (Fischer et al., 1988; Walter et al., 2000).
199 Several studies using ^{234}Th in the ACC confirm the high export around the Polar Front of the southern
200 Atlantic (Planchon et al., 2012; Rutgers van der Loeff et al., 2002; Usbeck et al., 2002), a situation also
201 observed in the South Pacific (Buesseler et al., 2001; Buesseler et al., 2003). Further south, in the
202 Weddell Gyre, production is thought to be limited by iron supply (Smetacek et al., 1997) and sediment
203 accumulation rates are very low. Nevertheless, Leynaert et al. (1993) observed in the Weddell Sea
204 biogenic silica production rates exceeding average rates in the ACC and Hoppema et al. (1999) showed
205 from a surface layer balance that export production in the central Weddell Gyre is larger than the
206 average for the Southern Ocean. Other studies point at blooms with high chlorophyll concentrations and
207 high export rates at these higher latitudes (Geibert et al., 2010). This discrepancy is explained by the
208 unusually shallow mineralization in the Gyre, as shown by ^{234}Th studies of Planchon et al. (2012) and
209 Usbeck et al. (2002). The first authors were able to quantify ^{234}Th excess and to relate it to
210 mineralization rates estimated from the distribution of particulate barite. They observed that the depth
211 range for mineralization was deeper in the region bounded by the Subantarctic Front and the APF (100-
212 600m) than in the (northern) Weddell Gyre (upper 400m). Usbeck et al. (2002) observed a ^{234}Th excess

213 in the depth range of just 100-200m in the Weddell Gyre, supporting the concept of an unusually
214 shallow depth of mineralization in this basin (Leynaert et al., 1993; Whitworth and Nowlin, 1987).

215 *Central Intermediate Water*

216 In the central Weddell Sea, the shallow mineralization gives rise to a water layer within the Warm Deep
217 Water characterized by low oxygen, high nitrate and total carbonate (TCO_2) (Hoppema et al., 2002;
218 Whitworth and Nowlin, 1987), coined Central Intermediate Water (CIW) by Whitworth and Nowlin
219 (1987). This water mass with at its core a neutral density around $27.88 \text{ kg}\cdot\text{m}^{-3}$ is derived from LCDW with
220 very low CFC content (Huhn et al., 2013), and is exposed to upwelling (Hoppema et al., 2002; Jullion et
221 al., 2014) and erosion from above by entrainment into surface waters. It exchanges isopycnally with
222 UCDW/LCDW in the north on a timescale of 3 years (Hoppema et al., 2002).

223 The shallow maximum in ^{210}Pb observed at GEOSECS station 89 (Somayajulu and Craig, 1976) (Fig. 4), in
224 the core of is now called CIW, was interpreted by Farley and Turekian (1990) to be produced locally, i.e.
225 by shallow mineralization and not by advection. Recently, Hoppema et al. (2015) confirmed that the
226 characteristics of the CIW described by Whitworth and Nowlin (1987) with its shallow nitrate maximum
227 indicating a maximum in remineralization rate at 200-500m depth are found consistently over many
228 years. They also found a corresponding maximum of silicate at much larger depth: 1000-2000m,
229 indicating that the opal skeletons dissolve at much greater depth than the organic matter. While this is a
230 usual phenomenon in many ocean areas it is somewhat surprising to find this deep opal dissolution in
231 the Weddell Gyre because Leynaert et al. (1993) had argued based on sediment trap data of Fischer et
232 al. (1988) that less than 1% of biogenic silica produced in the surface water of the northern Weddell Sea
233 reaches a depth of 800m.

234 In Fig. 2 we have extended the schematic meridional circulation model described by Chase et al. (2003)
235 to include the special situation in the Weddell Gyre with its upwelling, deep water formation, and

236 shallow mineralization in the Central Intermediate Water. In an upper meridional circulation cell,
237 nutrient-rich UCDW feeds the surface water and is then, on its way back north, depleted in silicate by
238 strong diatom production and correspondingly high opal flux just south of the APF. This flux also carries
239 ^{231}Pa to intermediate depth, preventing the ^{231}Pa to be carried north in surface waters. Instead, the cell
240 functions as a trap for ^{231}Pa , which settles out and accumulates at intermediate depth until it is exported
241 as WSDW and AABW. There are two reasons why this upper circulation cell is more effective in
242 accumulating ^{231}Pa than ^{230}Th : firstly, the strong affinity of Pa to opal, and secondly, the nonlinear depth
243 distribution of ^{231}Pa (and ^{210}Pb) north of the Polar Front. In those lower latitudes, ^{231}Pa and ^{210}Pb are
244 relatively constant from 1500m downwards, whereas ^{230}Th continues to increase linearly with depth.
245 The UCDW that upwells from mid-depth has approximately the same ^{231}Pa and ^{210}Pb activities but only
246 half the ^{230}Th activities if compared to water from full depth. In Fig.5, we have modified the model of
247 Rutgers van der Loeff and Berger (1993) of the nuclide activity profile in the Weddell Sea to take into
248 account the processes shown in Fig.2 that are essential aspects in the scavenging of ^{231}Pa and ^{210}Pb . The
249 modified model still includes a renewal of WSDW by LCDW on a time scale of 35 years, but is now
250 extended with the rapid exchange of WDW/CIW (100-500m) with UCDW/LCDW from N of Polar Front
251 and with the input of WSBW as described above (Fig. 3). The composition of recently produced WSBW is
252 derived in analogy to the procedure followed for ^{230}Th . For WDW we find ^{231}Pa and ^{210}Pb concentrations
253 of 0.35 and 140 dpm m^{-3} , and for recently formed WSBW concentrations of 0.23 and 89 dpm m^{-3} ,
254 respectively. Upwelling is set at 33m yr^{-1} (Hoppema et al., 1999), an upwelling rate that, within the large
255 errors in the budget for surface water layer, is supported by the analysis of Jullion et al. (2014). The
256 composition in the northern box is assumed to be set by the circumpolar current and is not changed in
257 the model.

258 The Th data are well represented if the scavenging process is described by a constant particle flux and
259 reversible scavenging (as in Rutgers van der Loeff and Berger, 1993) (Fig. 4). The modified model gives a

260 better representation of the ^{231}Pa and ^{210}Pb profiles than the original model, provided we allow the
261 fractionation factor to change with depth. The fractionation factors in deep waters are constrained by
262 the nuclide ratios observed in surface sediments (Table S2). We find the best fit with observations for an
263 8 times higher scavenging rate in surface waters for both Pa and Pb, decreasing with depth with a
264 relaxation depth of 2000m and 250m, respectively (Fig. 4). All model parameter values are listed in Table
265 S2. These three different scavenging depth regimes seem to be related to a combination of different
266 particle fluxes: a component with shallow mineralization that releases ^{210}Pb at shallow depth, an opal
267 flux that carries ^{231}Pa to depth where it is released upon opal dissolution (Hoppema et al., 2015), and a
268 third component that is not mineralized and sinks to the seafloor. The shallow release of ^{210}Pb points at
269 a relationship with the mineralization of organic matter. The strong increase in the Th/Pa fractionation
270 factor with depth (here from 0.36 to 2.9) has also been applied in 2D-scavenging models of the Atlantic
271 MOC (Lippold et al., 2012; Luo et al., 2010) and is in agreement with other data from the Southern
272 Ocean. Venchiarutti et al. (2011a) reported reduced F ratios in the surface 500m in the Drake Passage,
273 and from sediment trap results Chase et al. (2003) found a higher F ratio at depth than in surface
274 waters. F ratios calculated using $^{231}\text{Pa}/^{230}\text{Th}$ ratios in surface sediments increase with depth off
275 Southwest Africa (Scholten et al., 2008). However, using suspended particles, these latter authors found
276 a broad maximum in fractionation factor for the water column between $\sim 1000\text{m}$ and 4400m off
277 Southwest Africa, which fits with the observations of Moran et al. (2002) in the Argentine Basin.
278 Conversely, Hayes et al. (2015b) observed an opposite depth dependence in the North Atlantic Ocean, a
279 difference that is probably due to the minor role played by opal in their study.

280 **A mass balance of ^{230}Th and ^{231}Pa in the Atlantic sector of the Southern Ocean**

281 Further constraints on the ^{231}Pa and ^{230}Th activities in deep water leaving the Weddell Gyre as
282 WSDW/AABW can be based on mass balance considerations. A mass balance for ^{230}Th and ^{231}Pa for the

283 Southern Ocean presented by Chase et al. (2003) describes the fluxes of these nuclides across the APF
284 for all three ocean basins. In the Atlantic sector of the Southern Ocean, the region south of the APF
285 consists of two widely different zones: the northern part, between the APF and the Southern Boundary,
286 is part of the ACC with large export production, high diatom production and opal sedimentation (the
287 opal belt). South of the Southern Boundary we find the Weddell Gyre with low flux and low burial. In
288 order to make a mass balance of the Atlantic sector we refine the mass balance presented by Chase et
289 al. (2003) by separating the ACC from the Weddell Gyre.

290 Our analysis is based on the circulation of the Atlantic Sector of the Southern Ocean as given by Sloyan
291 and Rintoul (2001). From their analysis of hydrographic data (cruise tracks shown in Fig. 1) we use the
292 meridional fluxes across the SAVE 4 and Wedsea transects, the net zonal fluxes to the Indian (across
293 AJAX transect) and Pacific (across Drake Passage) Oceans, and the diapycnal fluxes across the neutral
294 density surfaces 27.4, 28.0 and 28.2 kg.m⁻³ (Sloyan and Rintoul, 2001, their Fig. 13, Table 1). The Wedsea
295 transect of the AJAX expedition separates the ACC from the Weddell Sea. Total volumes defined by the
296 sections were calculated with a matlab program using the published coordinates of the sections (Ajax,
297 DrakeP and SAVE4) for the area and the ETOPO05 database (NOAA, 1988) for the depth. Nuclide fluxes
298 for each water mass are calculated as the product of the mass flux and the literature value of their
299 nuclide activities (Table 2).

300 **Mass balance of Weddell Gyre**

301 We start with a mass balance of the Weddell Gyre. In the density range <28.0 kg.m⁻³ (Table 1) a
302 southward transport of about 1 Sverdrup (10⁶ m³ s⁻¹, Sv) of upwelling NADW is balanced by a northward
303 transport of surface water (AASW). Due to the difference in activity of the nuclides this implies a net
304 southward transport of 0.2±0.2 x 10⁶ dpm s⁻¹ of both ²³⁰Th and ²³¹Pa. As a result of the strong
305 accumulation of ²³⁰Th and ²³¹Pa in deep waters in the Weddell Gyre ($\gamma^n > 28.0 \text{ kg.m}^{-3}$) the outflowing

306 deep waters have higher concentrations of ^{230}Th and ^{231}Pa than the inflowing waters. Simple mass
307 balance requires that the cumulative nuclide flux in inflowing waters with concentration A_{in} plus in situ
308 production P is balanced by outflowing waters with concentration A_{out} plus burial B .

$$309 \quad \Sigma F(A_{\text{in}} - A_{\text{out}}) + P = B$$

310 The production of ^{230}Th in the Weddell Gyre is given by

$$311 \quad P_{230} = \lambda_{230} V_{\text{Weddell}}$$

312 and similarly for ^{231}Pa

$$313 \quad P_{231} = \lambda_{231} V_{\text{Weddell}}$$

314 Where λ_{230} and λ_{231} are the decay constants of ^{230}Th and ^{231}Pa . With a water volume V_{Weddell} in the
315 Weddell Gyre south of the Wedsea transect (Fig. 1) of $1.22 \times 10^{16} \text{ m}^3$, we thus have a direct relationship
316 between burial/production (B/P) ratio, the water flow F and the increase in concentration A .

317 Sloyan and Rintoul (2001) described the exchange through the Wedsea transect as a southward
318 transport of 11 Sv of deep waters consisting of 4 Sv in the density range LCDW ($28.0\text{-}28.2 \text{ kg}\cdot\text{m}^{-3}$) and 7
319 Sv with density $>28.2 \text{ kg}\cdot\text{m}^{-3}$ balanced by a northward transport of 11 Sv of WSDW/AABW ($\sigma^n >28.2 \text{ kg}\cdot\text{m}^{-3}$).
320 The estimates of the net production rate of WSDW/AABW in the Weddell Gyre vary widely. More
321 recently, Jullion et al. (2014) mentioned an export of AABW to the north of 8 ± 2 Sv of which 6 ± 2 Sv is
322 produced in the Weddell Gyre, the remainder being imported from the east. In a first analysis we
323 disregard this input from the Indian Ocean (see arguments below) and describe the deep water
324 circulation of the Weddell Gyre as an input of 8 Sv in the LCDW density range and an export of 8 Sv as
325 WSDW.

326 Following the analysis of Chase et al. (2003), we define characteristic concentrations for the water
327 masses (Table 2). LCDW flowing south has ^{230}Th and ^{231}Pa activities of 1.0 dpm m^{-3} and 0.35 dpm m^{-3}
328 (Rutgers van der Loeff and Berger, 1993). For the WSDW/AABW density range ($\gamma^n > 28.2 \text{ kg m}^{-3}$) we
329 distinguish three water types: the waters found in the western (WSDWW) and central/eastern (WSDWE)
330 Weddell Gyre and the AABW observed by Deng et al. (2014) in the Argentine Basin (AABWA), taken to
331 be representative for the AABW transport across SAVE4 (Table 2).

332 For the AABW/WSDW transport across the Weddell Sea transect, we consider two extreme options. Firstly,
333 in the perimeter and in the western part of the Weddell Gyre (WSDWW: Venchiarutti et al., 2011a, Sta
334 236) ^{230}Th and ^{231}Pa activities are 1.5 and 0.4 dpm m^{-3} , respectively, resulting in a net northward (export)
335 flux of 3.8 ± 4.0 and $0.2 \pm 1.1 \times 10^6 \text{ dpm} \cdot \text{s}^{-1}$ for ^{230}Th and ^{231}Pa respectively, and respective B/P ratio in the
336 Weddell Gyre of 0.62 and 0.78). However, if we take the high activities measured in the central Gyre
337 (WSDWE: 1.9 and 0.6 dpm m^{-3} for ^{230}Th and ^{231}Pa , respectively) to be representative for the 8 Sv of
338 WSDW flowing north, we find an export of 7.0 and $1.8 \times 10^6 \text{ dpm s}^{-1}$ with a B/P ratio in the Gyre of 0.3
339 and -0.9 , respectively. In the latter case, the export of ^{231}Pa exceeds the combined import and in situ
340 production, which in steady state is not possible. In other words, the northward outflowing
341 WSDW/AABW must have a lower ^{231}Pa concentration.

342 Inverting the argument, we can start with the observation of Walter et al. (2000) who found a B/P ratio
343 in the central Gyre of 0.4 for ^{230}Th . The $^{231}\text{Pa}/^{230}\text{Th}$ ratio on suspended particles and in surface sediments
344 far south of the APF is constant at 0.15 ± 0.03 (Walter et al., 1997) (Latitude $> 55^\circ\text{S}$, depth $> 1000\text{m}$,
345 <http://doi.pangaea.de/10.1594/PANGAEA.54596>) and the $^{231}\text{Pa}/^{230}\text{Th}$ production ratio is 0.093 , implying
346 that for ^{231}Pa the present B/P ratio is 0.65 . Using for ^{230}Th a B/P ratio of 0.4 ± 0.15 and for ^{231}Pa a B/P ratio
347 of 0.65 ± 0.15 , we find that a deep water flow of 8 Sv is enriched by $0.75 \pm 0.27 \text{ dpm m}^{-3}$ in ^{230}Th , a value in
348 agreement with the analysis of Rutgers van der Loeff and Berger (1993) (Sta 227) and within the range

349 of observations of Venchiarutti et al. (2011b)(Stations 131, 193). An analogous argument for ^{231}Pa shows
350 that the outflowing deep water is enriched by only 0.04 ± 0.02 dpm m^{-3} . This increase is small compared
351 to the range of activities observed in deep waters. The available data of the Weddell Gyre do not allow
352 to define the concentration in the outflow with sufficient accuracy to use them to calculate the net
353 outflow of ^{231}Pa to the north. The mass balance of the Weddell Gyre is a more appropriate way to define
354 the net northward loss of ^{231}Pa . Based on this mass balance consideration, we find that the export of
355 ^{230}Th and ^{231}Pa from the Weddell Gyre to the north is 6.0 ± 1.5 and $0.32\pm 0.14 \times 10^6$ dpm s^{-1} , respectively
356 (Fig. 6).

357 The above calculation showed that the high activity of ^{231}Pa observed in the central Weddell Sea (>0.6
358 dpm m^{-3} , Sta 131, 193) cannot be representative for the water masses leaving the Weddell Gyre as
359 WSDW/AABW. Indeed there are large horizontal gradients in the Weddell Gyre with higher CFC (Huhn et
360 al., 2013) and lower concentrations of Ba (Hoppema et al., 2010), ^{231}Pa (Venchiarutti et al., 2011b), TCO_2
361 (Hoppema et al., 2002) in the perimeter of the Gyre, and lower CFC and higher accumulation in the
362 center of the Gyre. We conclude that the exported water masses are derived from the outer reaches of
363 the gyre characterized by lower concentrations, whereas the central water masses have a longer
364 residence time allowing further accumulation of mineralization products (Ba, TCO_2) and isotopes
365 produced by in situ production (^{230}Th , ^{231}Pa).

366 This contrast is enhanced by the import from the Indian. The large input associated with the Antarctic
367 Slope Front (ASF), 14 Sv of CDW and 9 Sv of WSDW (Jullion et al., 2014) with relatively low ^{231}Pa
368 concentrations (Venchiarutti et al., 2011b, Sta 178) can only have very limited exchange with the high-
369 ^{231}Pa waters in the central Weddell Sea. If an appreciable part of this input from the East mixed into the
370 central waters of the gyre, the high ^{231}Pa observed there would imply an export far in excess of the
371 production rate. The Indian input must consequently largely be a through-flow, modified by its

372 interaction with Weddell Sea water masses, and exiting towards the ACC in the north (Jullion et al.,
373 2014).

374 **Mass balance of Atlantic sector of the ACC including the opal belt**

375 The mass balance of nuclides in the Atlantic sector of the ACC is calculated from the water transports
376 and nuclide concentrations in Tables 1 and 2, respectively. Our estimates of nuclide fluxes across SAVE 4
377 (Fig. 6) are somewhat lower than the estimates made by Deng et al. (2014) who gave a southward flux
378 of 6.35×10^6 dpm s^{-1} for ^{230}Th and 3.57×10^6 dpm s^{-1} for ^{231}Pa . Deng et al. (2014) based their estimate on
379 the hydrographical analysis of Vanicek and Siedler (2002) who use slightly different density limits of the
380 water masses and give different fluxes compared to Sloyan and Rintoul (2001). For the estimation of the
381 nuclide transport across the Wedsea transect, we use the mass balance considerations for the Weddell
382 Gyre south of the Wedsea transect as presented above.

383 The resulting activity balance for the entire water column (Fig. 6) shows that a net amount of 11.2×10^6
384 dpm s^{-1} of ^{230}Th and 1.8×10^6 dpm s^{-1} of ^{231}Pa is imported into the region between the hydrographic
385 sections SAVE 4, Wedsea, DrakeP and AJAX. This activity accumulates in the sediment together with the
386 activity produced within the area (3.1×10^7 and 2.9×10^6 dpm s^{-1} for ^{230}Th and ^{231}Pa , respectively,
387 assuming a volume of 3.81×10^{16} m^3). This results in a B/P ratio of 1.36 and 1.65 for ^{230}Th and ^{231}Pa ,
388 respectively (Fig. 6), which is in good agreement with an earlier estimate of the B/P ratio for ^{230}Th in the
389 Atlantic sector of the ACC in the range 1.12-1.42 (Walter et al., 2000). The calculated burial $^{231}\text{Pa}/^{230}\text{Th}$
390 activity ratio is 0.11 (Fig.6), well in excess of the production ratio of 0.093. In this area a strong
391 latitudinal gradient exists in $^{231}\text{Pa}/^{230}\text{Th}$ activity ratios in suspended particles and in surface sediments
392 from <0.093 north of the APF to >0.093 south of it (Walter et al., 1997). The much higher sediment rain
393 rates south of the APF (Geibert et al., 2005) explain why the average burial $^{231}\text{Pa}/^{230}\text{Th}$ activity ratio
394 exceeds the production ratio.

395 The net nuclide transport to the Indian and Pacific oceans has a large error because small concentration
396 changes in the ACC (130 Sv) between the Drake and Zero Meridian transects would cause a large source
397 or sink in the area. As already mentioned by Walter et al. (1997) it is difficult to estimate what part of
398 the additional nuclide sedimentation is exported to the Indian Ocean.

399 The budget of ^{231}Pa is strongly affected by the large southward transport of ^{231}Pa from the Argentine
400 basin (Deng et al., 2014; Yu et al., 1996), whereas the export from the Weddell Gyre is comparatively
401 small. The input of ^{231}Pa from the north causes a large excess sedimentation in the opal belt.

402 The low ^{230}Th activities found by Deng et al. (2014) in the AABW in the Argentine Basin make the
403 temperate South Atlantic also a source of ^{230}Th for the ACC. The export from the Weddell Gyre is of the
404 same magnitude as the supply from the north. If, as assumed in our analysis, all excess is deposited in
405 the Atlantic sector of the ACC and not transported to the Indian Ocean, the B/P ratio is 1.36 (Fig.6), at
406 the high end of the range for ^{230}Th sedimentation given by Henderson et al. (1999) and to be considered
407 when using ^{230}Th for flux normalization.

408 This mass balance in the Atlantic Ocean sector is very different from the circumpolar average fluxes
409 across the APF for which Chase et al. (2003) found an appreciable net southward transport of ^{231}Pa and a
410 relatively small net northward transport of ^{230}Th .

411

412 **Export of ^{231}Pa and ^{230}Th in AABW: indications of removal in bottom waters**

413 While WSBW ($\theta < -0.7^\circ\text{C}$, density $> 28.4 \text{ kg.m}^{-3}$) is locally depleted in ^{230}Th and ^{231}Pa as a result of deep
414 water formation (Fig. 3), this water mass is too dense to leave the Weddell Gyre. Yet, WSBW is mixed
415 vertically into WSDW and it is this overlying WSDW, strongly enriched in ^{230}Th and ^{231}Pa by accumulation
416 in the Weddell Gyre, that is the source of AABW leaving the Weddell Gyre northward and spreading out

417 over the deep basins in the Atlantic and Indian Oceans (Fig. 1). On its way north, it loses extreme
418 characteristics and becomes less dense by mixing with overlying waters of LCDW/WDW type.

419 We take as representative for the AABW export the nuclide composition at neutral density 28.2-28.3 kg
420 m^{-3} . We have argued above that the WSDW/AABW exported from the Weddell Gyre must have ^{230}Th
421 and ^{231}Pa activities 0.75 and 0.04 dpm m^{-3} higher than in the inflowing LCDW, which results in expected
422 activities of 1.3-1.8 and 0.35-0.39 dpm m^{-3} , respectively (boxes in Fig. 8). However, we do not find these
423 high ^{230}Th and ^{231}Pa activities in AABW north of the APF (Fig. 8). In the AABW density range Deng et al.
424 (2014) found ^{230}Th and ^{231}Pa activities of 0.83 ± 0.2 and $0.28 \pm 0.03 \text{ dpm m}^{-3}$, respectively. In the South
425 Sandwich Trench (Sta 1785) and in a 5160m deep basin east of this (Sta 1782), stations in an area of
426 outflow of AABW (Orsi et al., 1999), Rutgers van der Loeff and Berger (1993) observed dense waters
427 with in the density range $28.3 < \gamma_n < 28.4 \text{ kg.m}^{-3}$ activities much lower than at corresponding density in the
428 Weddell Gyre (Stations 131, 193, 227), while in the AABW density range $28.2 < \gamma_n < 28.3 \text{ kg.m}^{-3}$ ^{230}Th and
429 ^{231}Pa activities did not exceed 1.08 and 0.35 dpm m^{-3} , respectively (Fig. 8). The densest water sampled
430 north of the APF on the Zero Meridian had a neutral density of 28.31 kg m^{-3} (in 1989) and 28.23 kg m^{-3}
431 (in 2008) (Fig. 8). Between 40-53°S, dissolved ^{230}Th activities of 0.67-1.07 and dissolved ^{231}Pa activities of
432 0.28-0.35 dpm m^{-3} were found in waters with a density exceeding 28.2 kg m^{-3} (Fig. 8). In the SW Indian
433 Ocean, Thomas et al. (2006) found 1.03 ± 0.05 (1 SE) dpm m^{-3} dissolved ^{230}Th ($1.13 \pm 0.06 \text{ dpm m}^{-3}$
434 unfiltered) at neutral density 28.22 kg.m^{-3} (at 32.6°S, WIND3) (Fig. 8). Their southernmost ^{231}Pa data
435 (unfiltered) at AABW density are from 20°S (WIND15, 4400m, $\gamma_n = 28.26 \text{ kg m}^{-3}$) and were as low as 0.19
436 $\pm 0.01 \text{ dpm m}^{-3}$. The high activities found by Scholten et al. (2008) further north in the deep Cape Basin
437 appear not to be connected. They were found in water masses with neutral density $< 28.2 \text{ kg m}^{-3}$ (Fig. 8)
438 and cannot be directly derived from WSDW. Deng et al. (2014) noticed that their depth profiles of ^{230}Th
439 and ^{231}Pa did not continue to increase linearly with depth until the seafloor and argued that the
440 observed near-bottom depletion reflects a removal process at or near the seafloor. A comparison with

441 the expected outflow concentrations of AABW at this density (Fig. 8) shows that an even much larger
442 nuclide removal must take place during transit of AABW from the Weddell Gyre to the latitudes N of the
443 ACC and into the Argentine Basin. This supports the observation of Thomas et al. (2006) who reported a
444 decrease of ^{231}Pa with depth below the NADW layer in the SW Indian Ocean and speculated on a
445 removal of ^{231}Pa in deep waters while they cross the opal-rich ACC.

446 The northern edge of the Weddell Gyre is characterized by mountain ranges rising to depths of approx.
447 3000m. Only a few deeper passages in the Scotia Ridge (Orkney, Bruce and Discovery Passages) and
448 further East (South Sandwich Trench) allow concentrated outflow of AABW (Fig. 1). At other longitudes,
449 there may well be north-south exchange over the mountain ranges. We know from nephelometer data
450 (Biscaye and Eitrem, 1977) that below approx. 2500m the suspended load increases with depth. The
451 strong currents associated with the ACC fronts continue to full depth and maintain a nepheloid layer
452 over the seafloor, both at abyssal depths and over the ridges.

453 A benthic nepheloid layer was observed with transmissometry at all stations when we crossed the
454 America Antarctic Ridge during the Zero and Drake expedition (Fig. S1). The transmission values in the
455 benthic nepheloid layer were lower and consequently the particle load was higher at the northern side
456 of the Ridge. In this same area a similar nepheloid layer was observed throughout during the EIFEX
457 project, carried out in 2004 (Fig. S2). The ridges are situated within the opal belt (Geibert et al., 2005)
458 which means that they are covered with opal-rich sediments. Dissolved silicate concentrations in the
459 deepest waters just over the mountains are exceptionally high (Fig. S3), which could be due to diffusion
460 from the sediments but is more likely due to opal dissolution in the suspended phase.

461 Kretschmer et al. (2011) investigated grain-size dependent adsorption of ^{231}Pa and ^{230}Th to sediments
462 from the opal belt (Shona Ridge, PS1768-8, 52.5930°S, 4.4760°E, 3299 m) and showed that the easily
463 suspended fine fraction (<20 μm) and especially the diatom-rich slowly sinking coarser fraction (20-

464 50 μ m) had a high $^{231}\text{Pa}/^{230}\text{Th}$ ratio. Therefore, we conclude that the deep waters leaving the Weddell
465 Gyre over these ridges are exposed to strong interaction with suspended opal and lose part of the ^{231}Pa
466 and ^{230}Th that had accumulated in the Weddell Gyre. As a result of the low Pa/Th fractionation of opal,
467 this additional bottom scavenging is relatively less effective for ^{230}Th .

468 An additional removal likely takes place by contact of bottom waters with hydrothermal inputs.
469 Manganese (hydr)oxides formed around vent sites are very efficient in removing Th and Pa from the
470 water column (Hayes et al., 2015a). We have no direct evidence for hydrothermal scavenging but many
471 active vents have been observed along the Scotia Ridge and especially in the area of throughflow
472 towards the Argentine basin (German et al., 2000). Venting is also apparent on the Bouvet Triple
473 junction region near Sta 113 (Middag et al., 2011, Fig. S3) and on the Southwest Indian Ridge (Tao et al.,
474 2012). With a Th/Pa fractionation factor of 5.5 ± 2.8 for manganese (hydr)oxides (Hayes et al., 2015b),
475 hydrothermal inputs cause a preferential scavenging of Th over Pa, in contrast to opal scavenging with a
476 fractionation factor $F \leq 1$.

477

478 **A double ^{231}Pa and ^{230}Th trap**

479 The upper meridional circulation provides a ^{231}Pa trap in the Weddell Gyre that is closely related to the
480 nutrient trap that accumulates nutrients south of the Antarctic Polar Front and prevents their export in
481 northward flowing mode and intermediate waters (Chase et al., 2003). Part of the diatoms with their
482 adsorbed ^{231}Pa sink down to a lower circulation cell. The essentially complete dissolution of these
483 sinking opal particles in the deep Weddell Sea causes a release of the ^{231}Pa (Fig 4). However, in contrast
484 to silicate, which is exported in high concentrations in the deep AABW, ^{231}Pa is once again stripped from
485 the water column when deep waters are flowing northward through the opal belt. Here the high
486 dissolved ^{231}Pa activities come once again into contact with resuspended opal (see conceptual sketch in

487 Fig. 2). As a result, the meridional circulation in the Atlantic sector of the Southern Ocean provides a
488 double ^{231}Pa and ^{230}Th trap, accumulating ^{231}Pa and ^{230}Th in the water column of the Weddell Gyre and in
489 sediments of the opal belt, and yielding low ^{231}Pa and ^{230}Th concentrations in newly exported AABW.

490

491 **Conclusions**

492 Weddell Sea Bottom Water (WSBW, $\gamma_n > 28.4 \text{ kg/m}^3$) is depleted in ^{230}Th and ^{231}Pa as a result of bottom
493 water formation but, in contrast to the NADW ventilation in the North Atlantic, this water mass is too
494 dense to leave the Weddell Sea.

495 In the Weddell Gyre, ^{230}Th and ^{231}Pa are removed from upwelling waters in an upper circulation cell and
496 accumulate in WSDW ($28.4 > \gamma_n > 28.2 \text{ kg/m}^3$). The release of ^{231}Pa is coupled with the near complete
497 dissolution of opal with maximum Si and ^{231}Pa signals around 2000m.

498 WSDW is the Atlantic source of AABW. A mass balance shows that water leaving the Gyre exports
499 $0.3 \pm 0.1 \times 10^6 \text{ dpm s}^{-1}$ of ^{231}Pa and $6.0 \pm 1.5 \times 10^6 \text{ dpm s}^{-1}$ of ^{230}Th , corresponding to an increase in activity
500 of 0.04 and 0.75 dpm m^{-3} , respectively compared to inflowing waters. These enhanced activities are not
501 found north of the ACC, and consequently they must be retained during transit of AABW through the
502 ACC. The near-bottom depletion of ^{230}Th and ^{231}Pa observed by Deng et al. (2014) and Thomas et al.
503 (2006) in AABW is much larger if compared with the source WSDW than if only compared with a linear
504 extrapolation of the concentration-depth profiles.

505 This removal is caused and characterized by contact of outflowing deep waters with resuspended opal
506 rich sediments in the opal belt and probably by additional scavenging by hydrothermal inputs. As a
507 result, newly formed AABW is preconditioned by strong scavenging with very little fractionation (low F-
508 factor). Dissolved ^{231}Pa and ^{230}Th activities are controlled by the balance between accumulation in the

509 Weddell Gyre and removal in the deep waters of the opal belt. If this balance would vary with the
510 glacial-interglacial changes in productivity, opal flux and the position of the opal belt (Kumar et al., 1995)
511 relative to the mountain ranges, it would be expected to affect the $^{231}\text{Pa}/^{230}\text{Th}$ signal recorded in deep
512 sediments in the South Atlantic, an effect that should be considered when this signal is interpreted in
513 terms of deep water flow.

514 In the meridional circulation, ^{231}Pa and ^{230}Th are removed twice in the opal belt: once in the upper
515 circulation cell by scavenging on particles exported from surface waters and once again when outflowing
516 deep water comes into contact with resuspended opal rich sediments.

517

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526

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718 Table 1. Water masses and fluxes (Sv) into and out of the indicated neutral density layers in the area
 719 confined by the transects SAVE 4, Wedsea, DrakeP and AJAX (after Sloyan and Rintoul, 2001). For
 720 AABW/WSDW we distinguish the deep water flowing into in the Argentine Basin (AABWA).

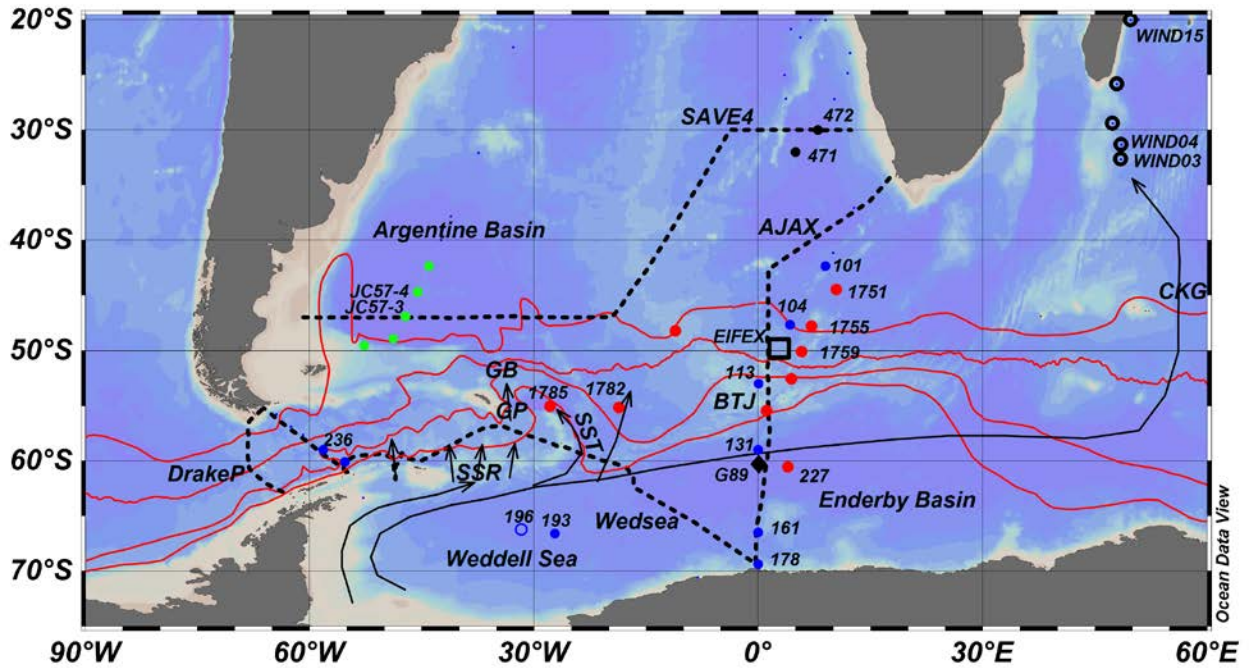
σ^{θ}	Across SAVE 4		Across Wedsea		Diapycnal				Pacific/Indian	
	in (S)	out (N)	in (N)	out (S)	above		below		in	out
<27.4		AASW 9.3±0.6						AASW 4	AASW 6.0±0.4	
27.4-28	NADW 6.7±1.4		AASW 1.0±0.2	NADW 1.0±0.2	AASW 4			NADW 1		NADW 2.2±1.1
28-28.2	LCDW 10.0±1.8			LCDW 8±2 *	NADW 1		WSDW 5			LCDW 11.9±1.9
>28.2		AABWA 6.6±1.3	WSDW 8±2 *			WSDW 5			WSDW 7.7±1.8	

721 * Jullion et al. (2014)

722 Table 2. Average activities (with ranges) of ^{230}Th and ^{231}Pa in the water masses. For AABW/WSDW we distinguish
 723 between the western (WSDWW) and central/eastern (WSDWE) part of the South Atlantic and the deep water
 724 flowing into in the Argentine Basin (AABWA)

	γ^n	^{230}Th	^{231}Pa	Reference
	$\text{kg}\cdot\text{m}^{-3}$	dpm m^{-3}	dpm m^{-3}	
AASW-AAIW	<27.4	0.2 ± 0.09 (0.17-0.36)	0.1 ± 0.06 (0.04-0.19)	(Venchiarutti et al., 2011b, Sta 101)
NADW	27.4-28	0.4 ± 0.09 (0.31-0.67*)	0.3 ± 0.14 (0.13-0.41)	(Venchiarutti et al., 2011b, Sts 101, 104)
LCDW	28-28.2	1 ± 0.1 (0.6-1.1)	0.35 ± 0.03 (0.31-0.35)	Rutgers van der Loeff and Berger (1993)
WSDWW	28.2-28.4	1.5 ± 0.2 (1.16-1.56)	0.4 ± 0.04 (0.37-0.44)	(Venchiarutti et al., 2011a, Sta 236)
WSDWE	28.2-28.4	1.9 ± 0.2 (1.61-1.95)	0.6 ± 0.06 (0.48-0.61)	(Venchiarutti et al., 2011b, Sta 193) (WS)
AABWA	≥ 28.2	0.83 ± 0.2	0.28 ± 0.03	Deng et al., (2014)

725 * leaving out datapoint (1245m) with exceptionally high particulate ^{230}Th



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Fig. 1. Map with published hydrographic sections used to make the budget (broken lines, SAVE4, AJAX ,

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Wedsea, DrakeP), stations of Rutgers van der Loeff and Berger (1993, red symbols), Deng et al. (2014, green

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symbols), Venchiarutti et al. (2011a,b, blue symbols), Huhn et al. (2013, open blue circle: station 196, data

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from 2008), Scholten et al. (2008, black dots), Thomas et al., (2006, black circles) and GEOSECS 89 (black

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diamond G89), and the location of the EIFEX study (black square) and the Bouvet Triple Junction (BTJ).

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Outflow pathways of AABW through four Passages in the South Scotia Ridge (SSR; from W to E: Philip,

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Orkney, Bruce and Discovery Passage), the Georgia Passage (GP) and Georgia Basin (GB), the South

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Sandwich Trench (SST) and the Crozet Kerguelen Gap (CKG) indicated by arrows. Red lines indicate average

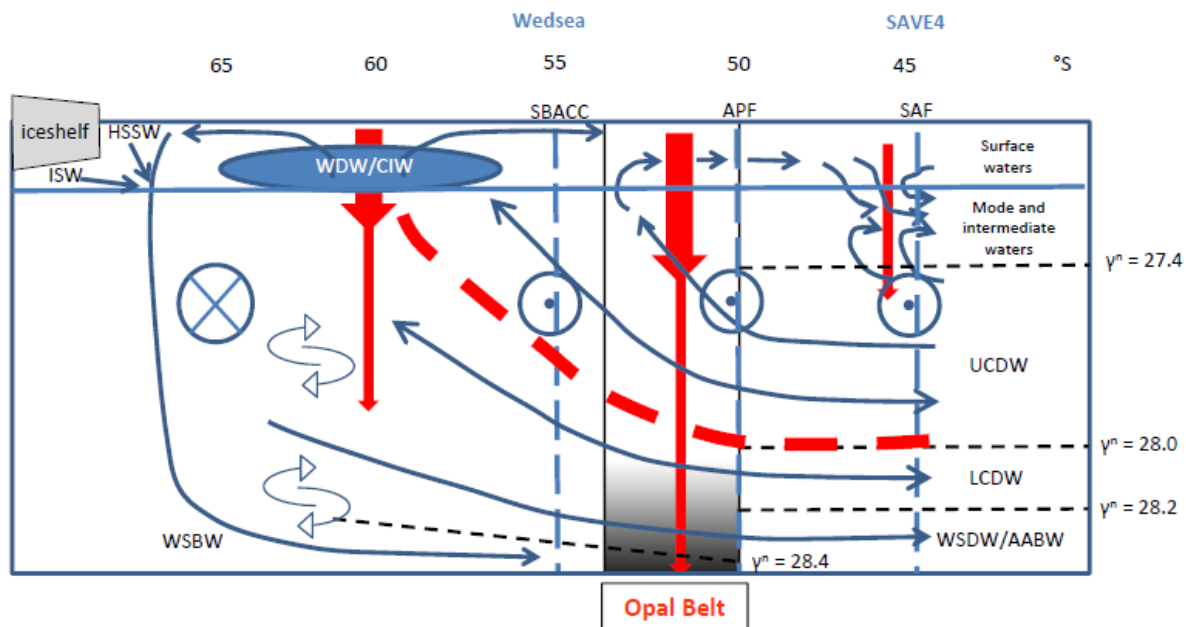
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position of (from N to S) Subantarctic Front, Polar Front, Southern ACC Front and ACC Southern Boundary

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(Orsi et al., 1995).

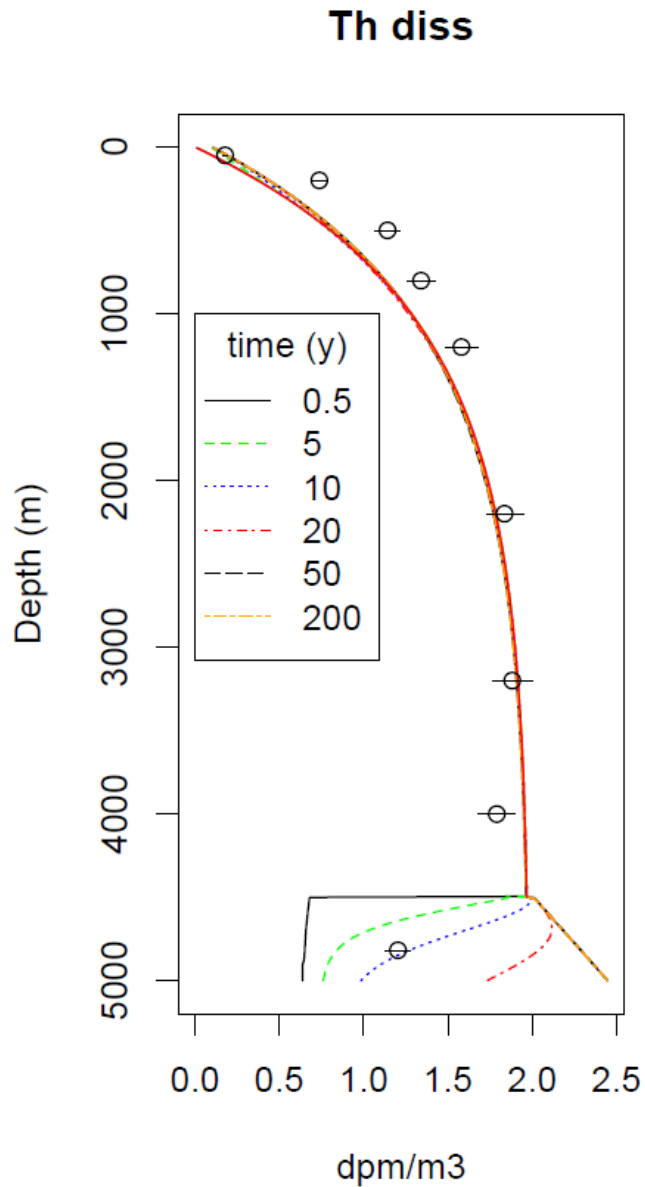
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739 Fig. 2. Conceptual diagram of 2-D meridional circulation of the Southern Ocean after Chase et al. (2003),
 740 modified to include the Weddell Gyre. The figure shows the approximate positions of the Weddell Sea and
 741 SAVE4 sections, upper and lower circulation cells separated by the dashed red line, eastward transport
 742 (circles with dots) in the ACC fronts, westward transport (circle with cross) from Indian Ocean in the
 743 Antarctic Slope Front (ASF) and in-between the Central Intermediate Water (WDW/CIW, blue area).
 744 Particle fluxes are indicated by red arrows, vertical mixing and isopycnal exchange by blue arrows. The
 745 high opal-rich deep particle flux in the productive zone south of APF (opal belt with nepheloid layer) is
 746 contrasted with the lower production zones further north and the zone with shallower mineralization in
 747 the Weddell Gyre. Near the ice shelf the production of High Salinity Shelf water (HSSW) and Ice Shelf
 748 Water (ISW) that mix with WDW to form WSBW is indicated.

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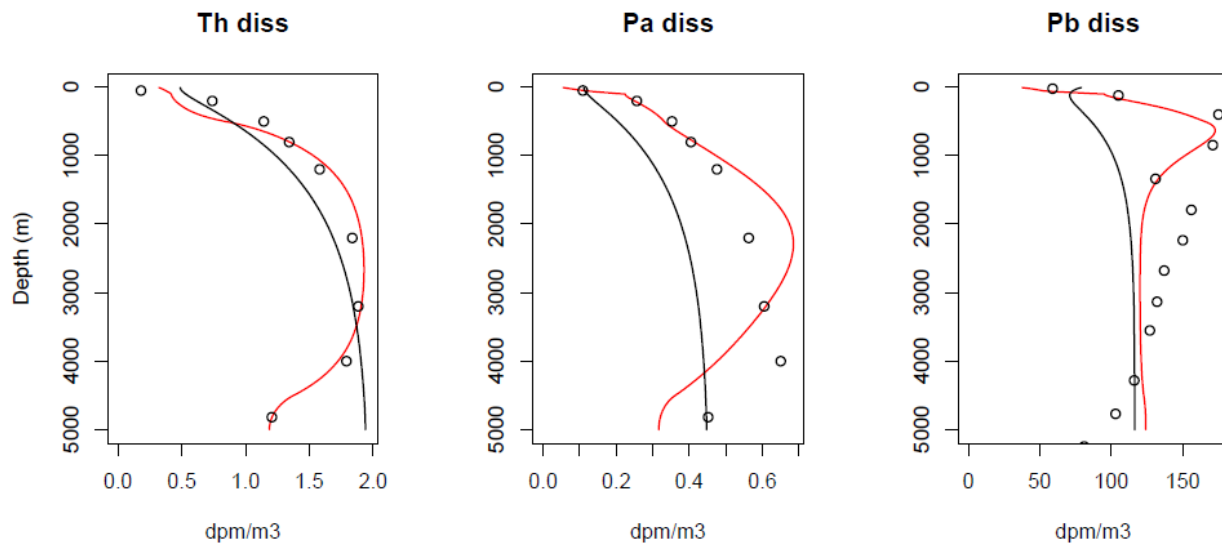
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Fig. 3. Distribution of dissolved ^{230}Th ($\pm\text{SE}$) at station PS71/193 (Venchiarutti et al., 2011b) with for depth range 0-4500m the steady-state model (analytical solution in red) of Rutgers van der Loeff and Berger (1993). Below 4500m dissolved ^{230}Th follows a time-dependent model. It starts at $t=0$ with newly formed WSBW (see text for estimate of ^{230}Th activity) and is then allowed to develop over time by ingrowth and reversible exchange with settling particles, showing that the value measured at 4800m is reached after about 10 years. This model run does not include eddy diffusion or upwelling in the bottom layer.



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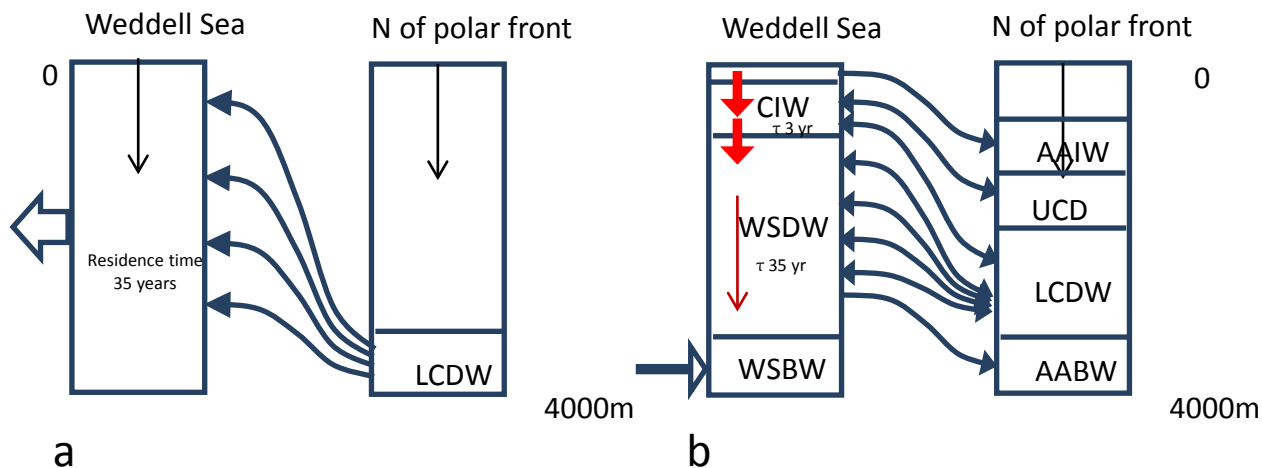
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Fig.4. Distribution of dissolved ^{231}Pa , ^{230}Th (Sta 193, Venchiarutti et al., 2011b), and of ^{210}Pb (Station GEOSECS 89, Somayajulu and Craig, 1976) in the Weddell Gyre (circles) compared with the model of Rutgers van der Loeff and Berger (1993) including vertical mixing and, for ^{210}Pb , atmospheric input (black lines). A modified model (red lines) includes ventilation of deep water by nuclide-depleted WSBW, and accumulation of nuclides in an upper meridional circulation cell by rapid exchange of WDW/CIW (100-500m) with UCDW/LCDW from N of Polar Front. For ^{231}Pa and ^{210}Pb , a reasonable fit with observations can only be obtained by assuming an increased scavenging of Pa and Pb relative to Th in surface waters, decreasing with depth with a relaxation depth of 2000m for ^{231}Pa and 250m for ^{210}Pb .

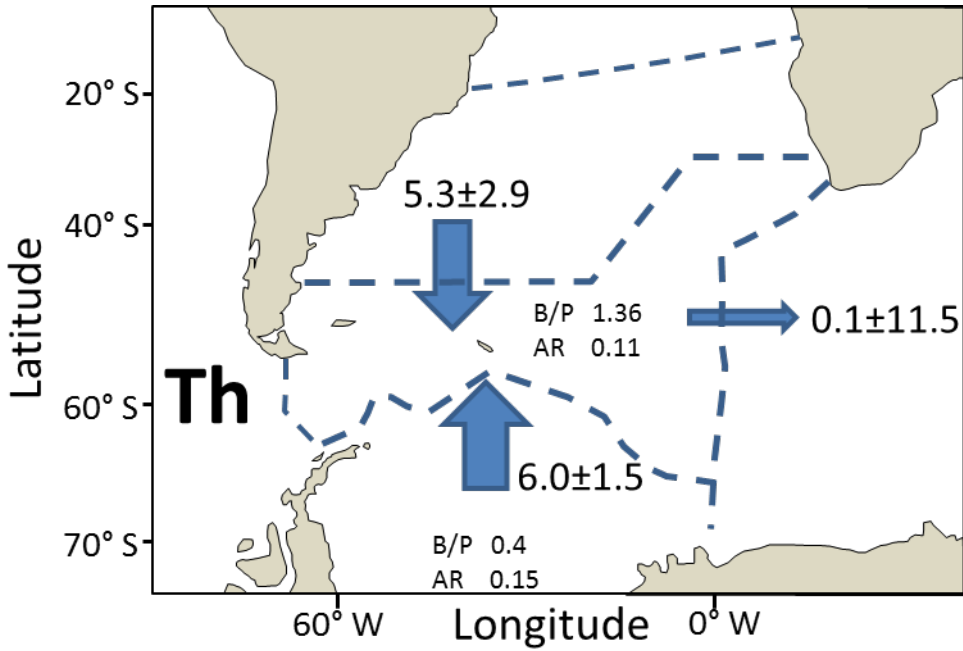
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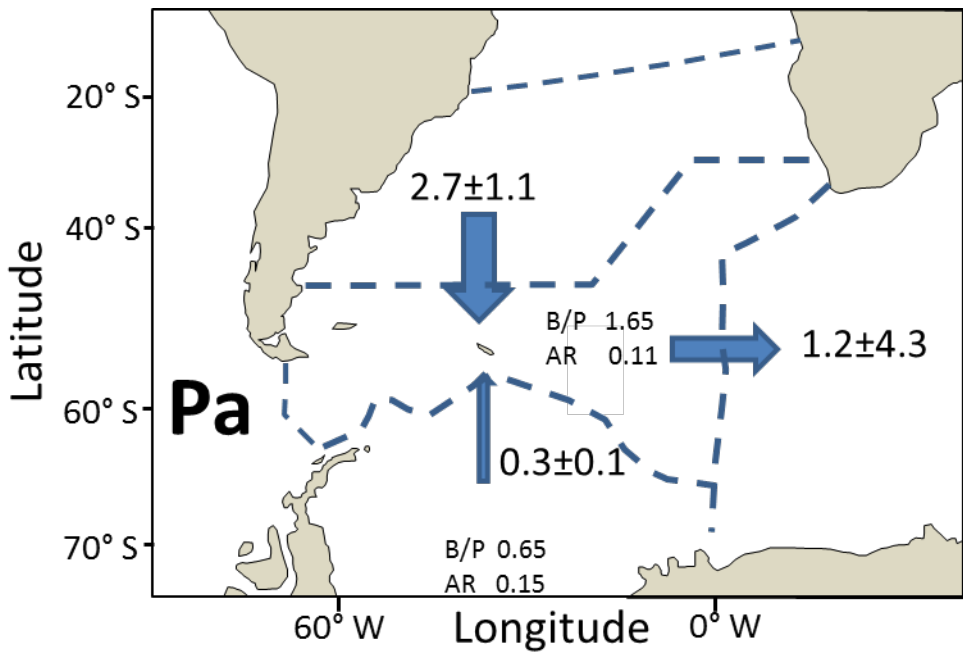
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769 Fig. 5. Model describing upwelling and scavenging in the Southern Ocean and adjacent Weddell Gyre. (a),
770 ²³⁰Th model of Rutgers van der Loeff and Berger (1993), considering an input by upwelling of LCDW in
771 entire water column of Weddell Gyre and a 60% reduced scavenging rate in Weddell Gyre. (b) modified
772 model (this paper) including ventilation of Weddell Sea by WSBW, rapid exchange of WDW/CIW (100-
773 500m) with UCDW/LCDW from N of Polar Front (Hoppema et al., 2002), and a scavenging term in the
774 Weddell Gyre that is constant for Th but depth-dependent for Pa and Pb.

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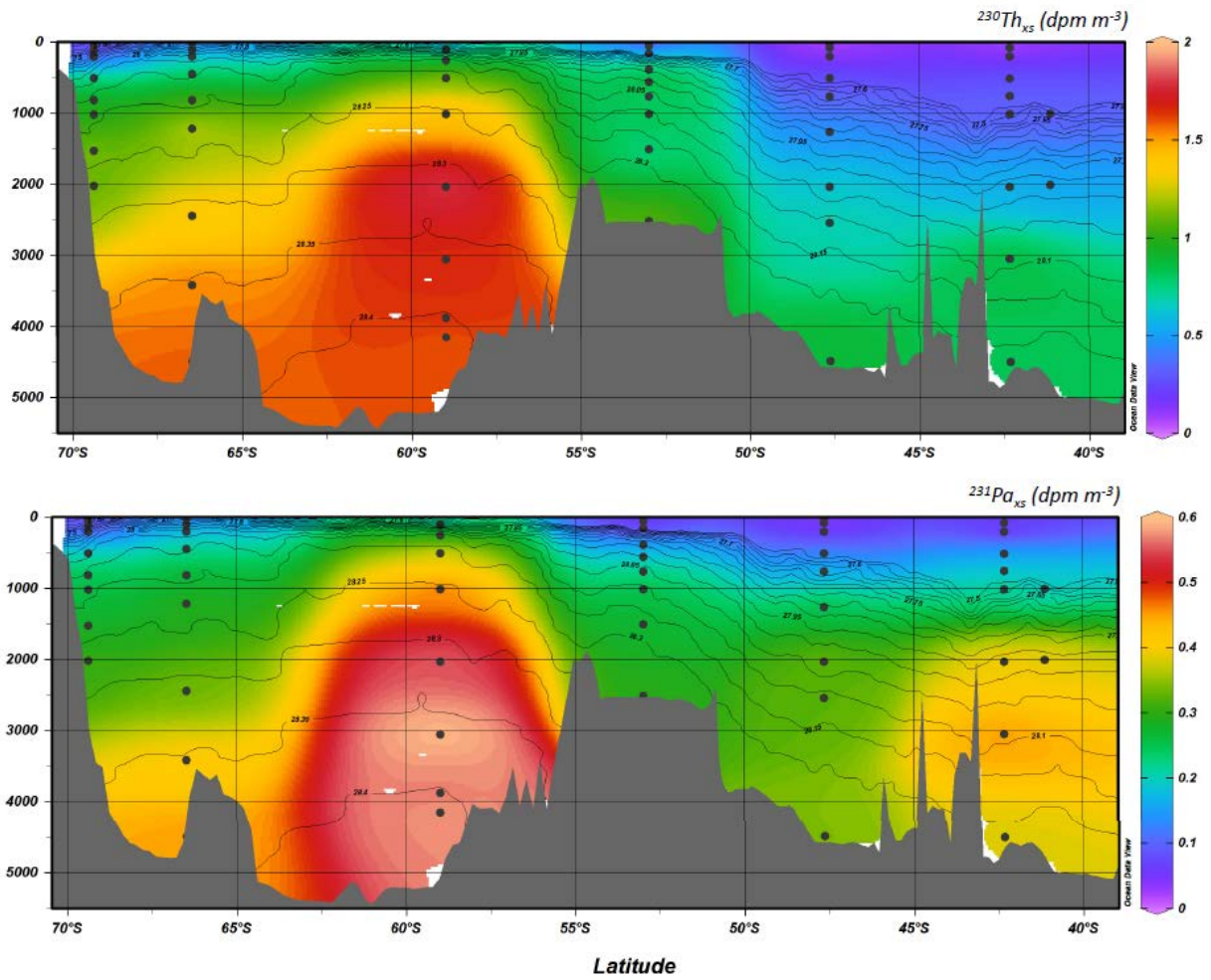
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778 Fig.6. Balance of ^{230}Th (a) and ^{231}Pa (b) activities based on circulation model of Sloyan and Rintoul (2001)

779 (Table 1) and radionuclide activities from Table 2. Fluxes from Weddell Sea based on production and

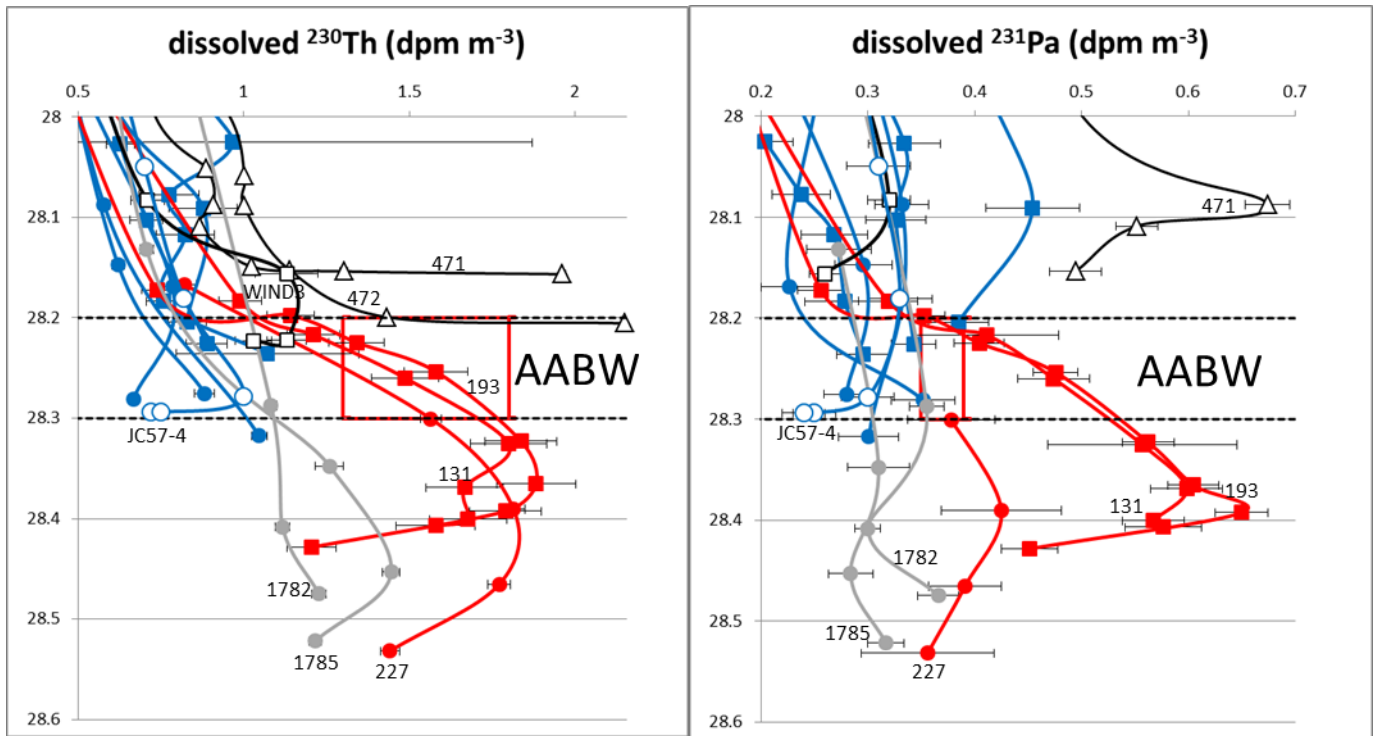
780 sedimentation constraints in Weddell Gyre. Arrows: Net fluxes with SE (10^6 dpm s^{-1}). B/P:

781 burial/production ratio. AR: Pa/Th burial activity ratio. Flux to Indian Ocean represents excess over input
782 from Pacific Ocean.



783
784 Fig.7 Vertical section along the Zero Meridian of excess dissolved ^{230}Th and ^{231}Pa (dpm/m^3 , from
785 Venchiarutti et al., 2011b) with contours of neutral density anomaly (kg m^{-3}) showing accumulation of
786 ^{230}Th and ^{231}Pa in the Weddell Gyre.

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Fig 8. Dissolved ^{230}Th and ^{231}Pa activities in the neutral density range (kg m^{-3}) of the AABW outflow.

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Observed activities (Deng et al., 2014, open circles; Rutgers van der Loeff and Berger, 1993, filled circles;

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Vencharutti et al., 2011b, filled squares) north of the APF (blue) are much lower than activities observed in

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the Weddell Gyre (red) and those expected for outflowing AABW based on the mass balance of the Weddell

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Gyre (boxes as described in text). Black symbols and lines represent observations in the Cape Basin

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(Scholten et al., 2008, open triangles; sta 472 refers to total concentrations) and the SW Indian Ocean

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(Thomas et al., 2006, open squares). Grey symbols are profiles observed close to the APF at stations 1782

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and 1785 (Fig. 1, Rutgers van der Loeff and Berger, 1993).

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