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# Variability and stability of anthropogenic CO<sub>2</sub> in Antarctic Bottom Waters observed in the Indian sector of the Southern Ocean, 1978-2018

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## Abstract

Antarctic bottom waters (AABWs) are known as a long term sink for anthropogenic CO<sub>2</sub> (C<sub>ant</sub>) but is hardly quantified because of the scarcity of the observations, specifically at an interannual scale. We present in this manuscript an original dataset combining 40 years of carbonate system observations in the Indian sector of the Southern Ocean (Enderby Basin) to evaluate and interpret the interannual variability of C<sub>ant</sub> in the AABW. This investigation is based on regular observations collected at the same location (63° E/56.5° S) in the frame of the French observatory OISO from 1998 to 2018 extended by GEOSECS and INDIGO observations (1978, 1985 and 1987).

At this location the main sources of AABW sampled is the fresh and younger Cape Darnley bottom water (CDBW) and the Weddell Sea deep water (WSDW). Our calculations reveal that C<sub>ant</sub> concentrations increased significantly in AABW, from about +7 μmol.kg<sup>-1</sup> in 1978-1987 to +13 μmol.kg<sup>-1</sup> in 2010-2018. This is comparable to previous estimates in other SO basins, with the exception of bottom waters close to their formation sites where C<sub>ant</sub> concentrations are about twice as large. Our analysis shows that the C<sub>T</sub> and C<sub>ant</sub> increasing rates in AABW are about the same over the period 1978-2018, and we conclude that the long-term change in C<sub>T</sub> is mainly due to the uptake of anthropogenic CO<sub>2</sub> in the different formation regions. This is however modulated by significant interannual to pluriannual variability associated with variations in hydrological (Θ, S) and biogeochemical (C<sub>T</sub>, A<sub>T</sub>, O<sub>2</sub>) properties. A surprising result is the apparent stability of C<sub>ant</sub> concentrations in recent years despite the increase in C<sub>T</sub> and the gradual acceleration of atmospheric CO<sub>2</sub>.

The C<sub>ant</sub> sequestration by AABWs is more variable than expected and depends on a complex combination of physical, chemical and biological processes at the formation sites and during the transit of the different AABWs. The interannual variability at play in AABW needs to be carefully considered on the extrapolated estimation of C<sub>ant</sub> sequestration based on sparse observations over several years.

## 1 Introduction

CO<sub>2</sub> atmospheric concentration has been increasing since the start of the industrialization (Keeling and Whorf, 2000). This increase leads to an ocean uptake of about a quarter of C<sub>ant</sub> emissions (Le Quéré et al., 2018; Gruber et al., 2019a). It is widely acknowledged that the Southern Ocean (SO) is responsible for 40 % of the C<sub>ant</sub> ocean sequestration (Gruber et al., 2009; Khatiwala et al., 2009; Matear, 2001; McNeil et al., 2003; Orr et al., 2001). Ocean C<sub>ant</sub> uptake and sequestration have the benefit to limit the atmospheric CO<sub>2</sub> increase but also result in a



37 gradual decrease of the ocean pH (Gattuso and Hansson, 2011; Jiang et al., 2019). Understanding the oceanic  $C_{\text{ant}}$   
38 sequestration and its variability is of major importance to predict future atmospheric  $\text{CO}_2$  concentrations, impact  
39 on the climate and impact of the pH change on marine ecosystems (de Baar, 1992; Orr et al., 2005; Ridgwell and  
40 Zeebe, 2005).

41  $C_{\text{ant}}$  in seawater cannot be measured directly and the evaluation of the relatively small  $C_{\text{ant}}$  signal from the total  
42 inorganic dissolved carbon ( $C_T$ ; around 3 %; Pardo et al., 2014) is still a challenge to overcome. Different  
43 approaches have been developed in the last 40 years to quantify  $C_{\text{ant}}$  concentrations in the oceans. The ‘historical’  
44 back calculation method based on  $C_T$  measurement and preformed inorganic carbon estimate ( $C^0$ ) was  
45 independently published by Brewer (1978) and Chen and Millero (1979). This method has been often applied at  
46 regional and basin scale (Chen, 1982, 1993; Goyet et al., 1998; Körtzinger et al., 1998, 1999; Poisson and Chen,  
47 1987; Lo Monaco et al., 2005a). More recently the TrOCA method (Tracer combining Oxygen, dissolved Carbon  
48 and total Alkalinity) has been developed (Touratier and Goyet, 2004a, 2004b; Touratier et al., 2007) and applied  
49 in various regions including the SO (e.g. Lo Monaco et al., 2005b; Sandrini et al., 2007; Pardo et al., 2014; Roden  
50 et al., 2016; Shadwick et al., 2014; Van Heuven et al., 2011; Kerr et al., 2018). Comparisons with other data-based  
51 methods show significant differences in  $C_{\text{ant}}$  concentrations, especially at high latitudes and more particularly in  
52 deep and bottom waters (Lo Monaco et al., 2005b; Vázquez-Rodríguez et al., 2009; Pardo et al., 2014). Thus, there  
53 is a need to better explore the  $C_T$  and  $C_{\text{ant}}$  temporal variability in the deep ocean, especially in the SO where  
54 observations are relatively sparse.

55 Antarctic bottom waters (AABWs) are of specific interest for the atmospheric  $\text{CO}_2$  and heat regulation as they  
56 play a major role in the meridional overturning circulation (Johnson et al., 2008; Marshall and Speer, 2012).  
57 AABWs represent a large volume of water by covering the majority of the bottom world ocean (Mantyla and Reid,  
58 1995), and their spreading in the interior ocean through circulation and water mixing (Siegenthaler and Sarmiento,  
59 1993) is a key mechanism for the long-term sequestration of  $C_{\text{ant}}$  and climate regulation. The AABW formation is  
60 a specific process occurring in few locations around the Antarctic continent (Orsi et al., 1999). In short, the AABW  
61 formation occurs when the Dense Shelf Water (DSW) flows down along the continental shelf. The DSW density  
62 required for this process to happen is reached by the increase in salinity ( $S$ ) due to brine release from the ice  
63 formation and by a decrease in temperature due to heat loss to either the ice-shelf or the atmosphere. Importantly,  
64 AABW formation process is enhanced by katabatic winds that open areas free of ice called polynyas (Williams et  
65 al., 2007). Indeed, katabatic winds are responsible for an intense cooling that enhance the formation of ice  
66 constantly pushed away by the wind, leading to cold and salty surface waters in contact with the atmosphere. The  
67 variable conditions of wind, ice production, surface water cooling and continental slope shape encountered around  
68 the Antarctic continent lead to different types of AABW, hence the AABW characteristics can be used to identify  
69 their formation sites.

70 The ability of AABW to accumulate  $C_{\text{ant}}$  has been controversial since one can believe that the ice coverage limits  
71 the invasion of  $C_{\text{ant}}$  in Antarctic surface waters (e.g. Poisson and Chen, 1987). This is however not the case in  
72 polynyas, and several studies have reported significant  $C_{\text{ant}}$  signals in AABW formation regions, likely due to the  
73 uptake of  $\text{CO}_2$  induced by high primary production (Roden et al., 2016; Sandrini et al., 2007; Shadwick et al.,  
74 2014; van Heuven et al., 2011, 2014). However, little is known about the variability and evolution of  $\text{CO}_2$  fluxes  
75 in AABW formation regions, and since biological and physical processes are strongly impacted by seasonal and  
76 interannual climatic variations (Fukamachi et al., 2000; Gordon et al., 2010, 2015; Gruber et al., 2019b; McKee et



77 al., 2011), the amount of  $C_{\text{ant}}$  stored in the AABWs may be very variable, which could bias the estimates of  $C_{\text{ant}}$   
78 trends derived from data sets collected several years apart (e.g. Williams et al., 2015; Pardo et al., 2017; Murata et  
79 al., 2019).

80 In this context of potentially high variability in  $C_{\text{ant}}$  uptake at AABW formation sites, as well as in AABW export,  
81 circulation and mixing, we used repeated observations collected in the Indian sector of the Southern Ocean to  
82 explore the variability in  $C_{\text{ant}}$  and  $C_T$  in AABW and evaluate their evolution over the last 40 years.

## 83 2 Studied area

### 84 2.1 AABW samples during the last 40 years

85 Most of the data used in this study were obtained in the frame of the long-term observational project OISO (Ocean  
86 Indien Service d'Observations) conducted since 1998 onboard the R.S.V. Marion-Dufresne (IPEV/TAAF). During  
87 these cruises, several stations are visited, but only one station is sampled down to the bottom (4800 m) south of  
88 the Polar Front at 63.0° E and 56.5° S (hereafter noted OISO-ST11). This station is located in the Enderby Basin  
89 on the Western side of the Kerguelen Plateau (Fig. 1) and coincides with the station 75 of the INDIGO-III cruise  
90 (1987). In our analysis, we also included data from the station 14 of the INDIGO-I cruise (1985) and the station  
91 430 of the GEOSECS cruise (1978) located near OISO-ST11 site (405 km and 465 km respectively). All the re-  
92 occupations used in this analysis are listed in Table 1.

93 Table 1.

### 94 2.2 AABWs circulation in the Atlantic and Indian sectors of the Southern Ocean

95 The circulation in the SO is mainly governed by the ACC that flows Eastward, while the Coastal Antarctic Current  
96 (CAC) flows Westward (Fig. 1). The ACC and the CAC influence the circulation of the entire water column,  
97 including the AABWs. The main AABW formation sites are the Weddell Sea where are produced deep and bottom  
98 waters (WSDW and WSBW, respectively; Gordon, 2001; Gordon et al., 2010), the Ross Sea (RSBW; Gordon et  
99 al., 2015, 2009), the Adelie Land coast (ALBW; Williams et al., 2008, 2010) and the Cape Darnley Polynya  
100 (CDBW; Ohshima et al., 2013). AABW formation in the Prydz Bay has also been observed (Rodehacke et al.,  
101 2007; Yabuki et al., 2006) from three polynyas and two ice shelves flowing into the Prydz Channel (Williams et  
102 al., 2016) and mixing with the CDBW. The CDBW and Prydz Bay bottom waters (hereafter called CDBW)  
103 represent a significant AABW export (13 % of all AABWs exports; Ohshima et al., 2013).

104 The largest bottom water source of the global ocean is the Weddell Sea (Gordon et al., 2001). The exported WSDW  
105 is a mixture of the Warm Deep Water (WDW) and the WSBW. The WSDW in the ACC and Weddell Gyre mixes  
106 with the Circumpolar Deep Water (CDW). A part of the WSDW deflecting southward with the ACC in the Enderby  
107 Basin reaches the Princess Elizabeth Trough (PET) region, East of the Kerguelen Plateau, where it mixes with  
108 other types of AABWs (Orsi et al., 1999).

109 In the PET sector, the CAC transports a mixture of RSBW and ALBW and accelerates Northward along the Eastern  
110 side of the Kerguelen Plateau (Mantyla and Reid, 1995; Fukamachi et al., 2010). Part of the ALBW-RSBW mixture  
111 also reaches the Western side of the Kerguelen Plateau (Orsi et al., 1999; Van Wijk and Rintoul, 2014) and mix  
112 with the CDBW. The mixture of CDBW and ALBW-RSBW either flows Westward with the CAC and dilutes with



113 the CDW (Meijers et al., 2010) or flow Northward (Ohshima et al., 2013) and mix with the older WSDW before  
114 reaching the location of our time-series station in the eastern Enderby Basin.  
115 Figure 1.

### 116 2.3 AABW definition

117 Nowadays, the distinction of water masses is usually performed according to neutral density ( $\gamma^n$ ) layers. In the SO,  
118 CDW and AABW properties are generally well defined in the range 28.15-28.27  $\text{kg.m}^{-3}$  and 28.27-bottom  
119 respectively (Orsi et al., 1999; Murata et al 2019). However, to interpret the long-term variability of the properties  
120 in the AABWs at our location, we prefer to adjust the AABW layer in a narrow band, and select the samples for  
121  $\gamma^n > 28.35 \text{ kg.m}^{-3}$  (range starting at 4200m to 4600m depending on the year, see Fig. 3).  $\gamma^n > 28.35 \text{ kg.m}^{-3}$   
122 corresponds to the AABW characteristics observed at higher latitudes in the Indian SO sector (Roden et al., 2016).

### 123 2.4 AABW composition at OISO-ST11

124 At each formation site, AABWs experienced significant temporal property changes, mostly recognized at decadal  
125 scale (e.g. freshening in the South Indian Ocean, Menezes et al., 2017) with potential impact on carbon uptake and  
126  $C_{\text{ant}}$  concentrations during AABW formation (Shadwick et al., 2013). The  $\Theta$ -S diagram constructed from yearly  
127 averaged data in bottom waters (Fig. 2) shows that the AABW at OISO-ST11 is a complex mixture of WSDW,  
128 CDBW, RSBW and ALBW. The coldest type of AABW was observed at the GEOSECS station at 60° S (-0.56  
129 °C), probably because it experienced less mixing with CDW compared to the warmer type of AABW observed at  
130 the INDIGO-1 station at 53° S (-0.44 °C). For the other cruises and years,  $\Theta$  in AABW ranges from -0.51 to -0.45  
131 °C with no clear indication on the specific AABW origin. The S range observed in the bottom waters at OISO-  
132 ST11 (34.65-34.67), illustrates either changes in mixing with various AABW sources or temporal variations at the  
133 formation site. Given the knowledge of deep and bottom waters circulation and characteristics (Figs 1 and 2) and  
134 the significant  $C_{\text{ant}}$  concentrations that we estimated at depth (Fig. 3), the main contribution at our location is likely  
135 the younger and colder CDBW for which relatively high  $C_{\text{ant}}$  concentrations have been recently documented  
136 (Roden et al., 2016). From its formation region, the CDBW can either flow westward with the CAC or flow  
137 northward in the Enderby Basin (Ohshima et al., 2013, Fig. 1). In the CAC branch, the CDBW mixes with the  
138 CDW along the Antarctic shelf and the continental slope between 80°E and 30°E (Meijers et al., 2010; Roden et  
139 al., 2016). On the western side of the Kerguelen Plateau, CDBW also mixes with RSBW and ALBW (Orsi et al.,  
140 1999; Van Wijk and Rintoul, 2014). In this context, the  $C_{\text{ant}}$  concentrations observed in the bottom layer at OISO-  
141 ST11 are probably not linked to a single AABW source, but are likely a complex interplay of AABW from different  
142 sources with different biogeochemical properties.

143 Figure 2.

## 144 3 Material and methods

### 145 3.1 Validation of the data

146 For 1998-2004, the OISO data were quality controlled in CARINA (Lo Monaco et al., 2010) and for 2005 and  
147 2009-2011 in GLODAPv2 (Key et al., 2015; Olsen et al., 2016, 2019). The 3 additional stations from GEOSECS  
148 and INDIGO were qualified in GLODAP-v1 (Key et al., 2004) and previously used for the first  $C_{\text{ant}}$  estimates in



149 the Indian Ocean (Sabine et al., 1999). The data for INDIGO III (1987) have been revisited in GLODAP-v2 but  
150 the correction applied on  $A_T$  values leads to a suspicious offset and we decided to use the adjustment proposed in  
151 GLODAP-v1 and confirmed in CARINA.

152 For the recent OISO cruises conducted in 2012-2018 not yet qualified in the GLODAP project, we have proceeded  
153 to a data control mainly based on repeated observations in deep waters (CDW) where  $C_{ant}$  concentrations are low  
154 and subject to very small changes from year to year. At this location, the seasonal variations of all properties are  
155 only observed in the mixed-layer, about 50 m in austral summer and 150 m in winter (Metzl et al., 2006). Therefore,  
156 for deep water analysis, we used the observations available for all seasons (Table 1).

### 157 **3.2 Biogeochemical measurements**

158 Measurement methods during OISO cruises were previously described (Jabaud-Jan et al., 2004; Metzl et al., 2006).  
159 In short, measurements were obtained using Conductivity-Temperature-Depth (CTD) casts fixed on a 24 bottles  
160 rosette equipped with 12 L General Oceanics Niskin bottles.  $\Theta$  (in °C) and S (no unit) measurements have an  
161 accuracy of 0.01 of their respective units.  $C_T$  and  $A_T$  were sampled in 500 mL glass bottles and poisoned with 100  
162  $\mu\text{L}$  of  $\text{HgCl}_2$  saturated solution to halt biological activity. Discrete  $C_T$  and  $A_T$  samples were analyzed onboard by  
163 potentiometric titration derived from the method developed by Edmond (1970) using a closed cell. The accuracy  
164 for  $C_T$  and  $A_T$  varies from 1 to 3.5  $\mu\text{mol.kg}^{-1}$  (depending on the cruise) and is determined by sample duplicates in  
165 surface, at 1000 m and bottom waters. All measurements were calibrated with Certified Reference Materials  
166 (CRMs) provided by A.G. Dickson laboratory (Scripps Institute of Oceanography).  $\text{O}_2$  was determined by an  
167 oxygen sensor fixed on the rosette. These values were adjusted using measurements obtained by Winkler titrations  
168 using a potentiometric titration system (at least 12 measurements for each profile). Thiosulphate solution used in  
169 Winkler titration was calibrated using iodate standard solution to provide the standard  $\text{O}_2$  accuracy of 2  $\mu\text{mol.kg}^{-1}$ .  
170 Nitrate ( $\text{NO}_3$ ) and Silicate (Si) were measured onboard or offshore with an automatic colorimetric Technicon  
171 analyser following the methods described by Tréguer and Le Corre (1975) until 2008, and the revised protocol  
172 described by Aminot and K erouel (2007) since 2009. Based on replicate measurements for deep samples we  
173 estimate an error of about 0.3 % for both nutrients.  $\text{NO}_3$  concentrations are not available for all the cruises used  
174 in this analysis. The mean  $\text{NO}_3$  concentrations in the AABW at OISO-ST11 is  $32.8 \pm 1.2 \mu\text{mol.kg}^{-1}$  while the  
175 average value derived from GLODAP-v2 database in bottom waters south of 50°S in the South Indian is  $32.4 \pm$   
176  $0.6 \mu\text{mol.kg}^{-1}$ . The lack of  $\text{NO}_3$  for few cruises has been palliated by considering a standard value of 33  $\mu\text{mol.kg}^{-1}$   
177 with a limited impact on  $C_{ant}$  determined by the  $C^\circ$  method (from 0.1  $\mu\text{mol.kg}^{-1}$  to 1.7  $\mu\text{mol.kg}^{-1}$  on the mean  
178 annual values).

### 179 **3.3 $C_{ant}$ calculation using the TrOCA method**

180 The TrOCA method was first presented by Touratier and Goyet (2004a, 2004b) and revised by Touratier et al.  
181 (2007). Following the concept of the quasi-conservative tracer NO (Broecker, 1974), TrOCA is a tracer defined as  
182 a combination of  $\text{O}_2$ ,  $C_T$  and  $A_T$ , following:

$$183 \text{TrOCA} = \text{O}_2 + a \left( C_T - \frac{1}{2} A_T \right), \quad (1)$$

184 where  $a$  is the Redfield ratio.



185 The temporal change in TrOCA is independent of biological processes and can be attributed to anthropogenic  
186 carbon (Touratier and Goyet, 2004a). Therefore,  $C_{ant}$  can be directly calculated from the difference between  
187 TrOCA and its pre-industrial value  $TrOCA^0$ :

$$188 \quad C_{ant} = \frac{TrOCA - TrOCA^0}{a}, \quad (2)$$

189 where  $TrOCA^0$  is evaluated as a function of  $\theta$  and  $A_T$  (Eq. 3):

$$190 \quad TrOCA^0 = e^{\left[ b - (c) \cdot \theta - \frac{d}{A_T^2} \right]}, \quad (3)$$

191 In these expressions, coefficients  $a$ ,  $b$ ,  $c$  and  $d$  were adjusted by Touratier et al. (2007) from free anthropogenic  
192  $CO_2$  deep waters using the tracers  $\Delta^{14}C$  and CFC-11 from the GLODAP-V1 database (Key et al., 2004). The final  
193 expression used to calculate  $C_{ant}$  is:

$$194 \quad C_{ant} = \frac{O_2 + 1.279 \left( C_T - \frac{1}{2} A_T \right) - e^{\left[ 7.511 - (1.087 \cdot 10^{-2}) \cdot \theta - \frac{7.81 \cdot 10^5}{A_T^2} \right]}}{1.279}, \quad (4)$$

195

196 The consideration of the errors on the different parameters involved in the TrOCA method results in an uncertainty  
197 of  $\pm 6.25 \mu\text{mol.kg}^{-1}$  (mostly due to the parameter  $a$ , leading to  $\pm 3.31 \mu\text{mol.kg}^{-1}$ ). As this error is relatively large  
198 compared to the expected  $C_{ant}$  concentrations in deep and bottom SO waters (Pardo et al., 2014) we will compare  
199 the TrOCA results using another indirect method to interpret  $C_{ant}$  changes over 40 years.

#### 200 **3.4 $C_{ant}$ calculation using the preformed inorganic carbon method**

201 To support the  $C_{ant}$  trend determined with the TrOCA method,  $C_{ant}$  was also estimated using a back-calculation  
202 approach noted  $C^0$  (Brewer, 1978; Chen and Millero, 1979), previously adapted for  $C_{ant}$  estimates along the  
203 WOCE-I6 section between Africa and Antarctica (Lo Monaco et al., 2005a). This method consists in the correction  
204 of the measured  $C_T$  for the biological contribution ( $C_{bio}$ ) and the preindustrial preformed  $C_T$  ( $C_{0,PI}$ ):

$$205 \quad C_{ant} = C_T - C_{bio} - C_{0,PI}, \quad (5)$$

206  $C_{bio}$  (Eq. 6) depends on carbonate dissolution and organic matter remineralization, taking account of the corrected  
207 Redfield ratio from Kortzinger et al. (2001):

$$208 \quad C_{bio} = 0.5 \Delta A_T - (C/O_2 + 0.5 N/O_2) \Delta O_2, \quad (6)$$

209 Where  $C/O_2 = 106/138$  and  $N/O_2 = 16/138$ .  $\Delta A_T$  and  $\Delta O_2$  are the difference between the measured values ( $A_T$  and  
210  $O_2$ ) and the preindustrial values ( $A_T^0$  and  $O_2^0$ ).  $A_T^0$  (Eq. 7) has been computed by Lo Monaco et al. (2005a) as a  
211 function of  $\Theta$ ,  $S$  and the conservative tracer  $PO$ :

$$212 \quad A_T^0 = 0.0685 PO + 59.79 S - 1.45 \Theta + 217.1, \quad (7)$$

213  $PO$  (Eq. 8) has been defined by Broecker (1974) and depends on the equilibrium of  $O_2$  with phosphate ( $PO_4$ ). When  
214  $PO_4$  data are not available, nitrate ( $NO_3$ ) can be used instead (Anderson and Sarmiento, 1994):

$$215 \quad PO = O_2 + 170 PO_4 = O_2 + 170/16 NO_3, \quad (8)$$

216 To determine  $O_2^0$ , it is assumed that the surface water is in full equilibrium with the atmosphere ( $O_2^0 = O_{2,sat}$ ; Benson  
217 and Krause, 1980) and after only impacted by the biological activity (Weiss, 1970). The correction of  $O_{2,sat}$   
218 has been proposed by Lo Monaco et al. (2005a) to take account of the undersaturation of  $O_2$  due to sea-ice cover.  $\Delta O_2$   
219 is, therefore, corrected by assuming a mean mixing ratio of the ice-covered surface waters  $k=50\%$  (Lo Monaco et  
220 al., 2005a), and a mean value for  $O_2$  undersaturation in ice-covered surface waters  $\alpha=12\%$  (Anderson et al., 1991)  
221 according to Eq. 9:



222  $\Delta O_2 = (1 - \alpha k)O_{2,sat} - O_2 = AOU$  , (9)

223  $C_{0,PI}$  in equation 5 is a function of the current performed  $C_T$  ( $C_{0,obs}$ ) and a reference water term (Eq. 10):

224  $C_{0,PI} = C_{0,obs} + [C_T - C_{bio} - C_{0,obs}]_{REF}$  , (10)

225 Where the reference water term has been computed using an optimum multiparametric (OMP) model and defined  
226 as  $51 \mu\text{mol.kg}^{-1}$  from North Atlantic deep water (Lo Monaco et al., 2005a) and  $C_{0,obs}$  has been computed similarly  
227 as  $A_T^0$  (Eq. 11):

228  $C_{0,obs} = -0.0439PO + 42.79S - 12.02\theta + 739.8$  , (11)

229 For more details in the  $C^0$  method, which has a final error of  $\pm 6 \mu\text{mol.kg}^{-1}$ , especially on the determination of  
230 reference water terms and on the errors of this method, please see Lo Monaco et al. (2005a).

#### 231 4. Results

232 The vertical distribution of hydrological and biogeochemical properties observed in deep and bottom waters and  
233 their evolution over the last 40 years are displayed in Fig. 3. The LCDW layer ( $\gamma^n = 28.15\text{-}28.27 \text{ kg.m}^{-3}$ ) is  
234 characterized by maximum AOU values (Fig. 3c), maximum  $C_T$  concentrations (Fig. 3d) and minimum  $C_{ant}$   
235 concentrations (Fig. 3a).  $C_{ant}$  concentrations were not significant in the LCDW until the end of the 1990s ( $<6$   
236  $\mu\text{mol.kg}^{-1}$ ), then our data show a sudden increase in  $C_{ant}$  between January and December 1998, followed by  
237 relatively constant  $C_{ant}$  concentrations ( $10 \pm 3 \mu\text{mol.kg}^{-1}$ ). In the core of AABW ( $\gamma^n > 28.35 \text{ kg.m}^{-3}$ ), well identified  
238 by low  $\theta$ , low S, high  $O_2$  and low AOU,  $C_{ant}$  concentrations are higher than in the overlying deep waters (Fig. 3a)  
239 and increased from  $5 \pm 4 \mu\text{mol.kg}^{-1}$  in 1978,  $7 \pm 4 \mu\text{mol.kg}^{-1}$  in the mid-1980s to  $13 \pm 2 \mu\text{mol.kg}^{-1}$  at the end of the  
240 1990s and up to  $19 \pm 2 \mu\text{mol.kg}^{-1}$  in 2004 (Fig. 4a). Figure 4a also shows a very good agreement between the TrOCA  
241 method and the  $C^0$  method for both the magnitude and variability of  $C_{ant}$  in the core of AABW. Our results show  
242 a mean  $C_{ant}$  trend in AABW of  $+1.6 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  over the full period and a maximum trend of the order of  
243  $+6.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  over 1987-2004 (Table 2). These trends are lower than the theoretical trend expected from  
244 the increase in atmospheric  $\text{CO}_2$ . Indeed, assuming that the surface ocean  $f\text{CO}_2$  follows the atmospheric growth  
245 rate ( $+1.8 \mu\text{atm.year}^{-1}$  over 1978-2018), the theoretical  $C_{ant}$  trend at the AABW formation sites would be of the  
246 order of  $+8 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ . The observed slow  $C_{ant}$  trends can be partly explained by the transit time for AABW  
247 to reach our study site and the mixing of AABW with older CDW waters that contain less  $C_{ant}$  (Fig. 3).

248 Figure 3.

249 To investigate changes in the accumulation of  $C_{ant}$  in AABW, Fig. 4 shows the evolution of  $C_T$ ,  $A_T$ ,  $O_2$ ,  $\theta$  and S  
250 (properties used to estimate  $C_{ant}$ ), as well as the “natural” component of  $C_T$  ( $C_{Tnat}$  calculated as the difference  
251 between  $C_T$  and  $C_{ant}$ ). Over the full period,  $C_T$  increased by  $2.0 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ , mostly due to the  
252 accumulation of  $C_{ant}$  (Table 2). Our data also show a significant decrease in  $O_2$  concentrations by  $0.8 \pm 0.4 \mu\text{mol.kg}^{-1}.$   
253  $\text{decade}^{-1}$  over the 40-years period (Fig. 4c) that could be caused by reduced ventilation, as suggested by  
254 Schmidtko et al. (2017) who observed significant  $O_2$  loss in the global ocean. In the deep Indian SO sector, these  
255 authors found a trend approaching  $-1 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  over 50 years (1960-2010), which is consistent with our  
256 data. We did not detect any significant trend in  $A_T$ ,  $\theta$  and S over the full period, but on shorter periods our data  
257 show a significant decrease in  $A_T$  from the mid-1980s to 2004 (Fig. 4d, Table 2) that is also observed in the  
258 overlying deep waters (Fig. 3f). This could suggest reduced calcification in the upper ocean leading to less sinking  
259 of calcium carbonate tests and hence a decrease in  $C_{Tnat}$  (i.e. for this period the increase in  $C_T$  was lower than the



260 accumulation of  $C_{\text{ant}}$ ). This event is followed by an increase in  $C_{\text{Tnat}}$  since 2004 associated to a rapid decrease in  
261  $O_2$  (increase in AOU) and a decrease in  $C_{\text{ant}}$  (Table 2). These recent trends were associated with a small increase  
262 in  $\theta$  (Fig. 4e, Table 2), but no significant trend in  $S$  (Fig. 4f). The increase in  $C_{\text{Tnat}}$  is thus unlikely originating  
263 solely from increased mixing with LCDW during bottom waters transport. Enhanced organic matter  
264 remineralization is also unlikely since nitrate did not show any significant trend (Table 2).

265 Table 2.

266 Figure 4.

267 Importantly, our data show substantial interannual variations in AABW properties, which could significantly  
268 impact the trends estimated from limited reoccupations (e.g. Williams et al., 2015; Pardo et al., 2017; Murata et  
269 al., 2019). For example, we found relatively higher  $C_{\text{ant}}$  concentrations in 1985 ( $10 \mu\text{mol.kg}^{-1}$ ) compared to 1978  
270 and 1987 ( $5 \mu\text{mol.kg}^{-1}$ ). This is linked to a signal of low  $S$  in 1985 that could be due to a larger contribution of  
271 fresher AABW or reduced mixing with saltier LCDW (Fig. 3h). Over the last decade (2009-2018), our data show  
272 large and rapid changes in  $S$  that are partly reflected on  $C_T$  and  $O_2$ , and that could explain the relatively low  $C_{\text{ant}}$   
273 concentrations observed over this period. Indeed, the  $S$  maximum observed in 2012 (correlated to higher  $\theta$ ) is  
274 associated with a marked  $C_T$  minimum (surprisingly almost as low as in 1987), as well as low  $A_T$  (hence low  $C_{\text{Tnat}}$ ),  
275 and low nitrate concentrations. These anomalies point to a change in AABW characteristics rather than a change  
276 in mixing with the underlying deep waters, and since they were associated with a decrease in  $C_{\text{ant}}$  concentrations,  
277 one may argue for an increased contribution of bottom waters ventilated far away from our study site (possibly  
278 from the Ross Sea due to higher  $S$ , Fig. 2). A few years later our data show a  $S$  minimum (correlated to lower  $\theta$ ),  
279 associated with a rapid increase in  $C_T$  and a rapid decrease in  $O_2$  between 2013 and 2016, suggesting the  
280 contribution of a closer AABW such as the CDBW. The freshening of  $-0.01$  in  $S$  that we observed on the Western  
281 side of the Kerguelen Plateau was also observed on the Eastern side of the Plateau by Menezes et al. (2017). In  
282 this region, Menezes et al. (2017) evaluated a change in salinity by about  $-0.008.\text{decade}^{-1}$  from 2007 to 2016  
283 (against  $-0.002.\text{decade}^{-1}$  between 1994 and 2007), suggesting an acceleration of the AABW freshening in recent  
284 years. However, they also reported a warming by  $+0.06 \text{ }^\circ\text{C}.\text{decade}^{-1}$ , while we observed cooler temperature in  
285 2016-2018. This suggests that we sampled a different mixture of AABWs.

## 286 5. Discussion

### 287 5.1. $C_{\text{ant}}$ concentrations

288 In order to compare our  $C_{\text{ant}}$  estimates with other studies, we separated the 40-years time-series into 3 periods: the  
289 first period (1978-1987) corresponds to historical data when  $C_{\text{ant}}$  is expected to be low; the second period (1998-  
290 2004) starts when the first OISO cruise was conducted (and using CRMs for  $A_T$ ,  $C_T$  measurements) and lasts when  
291  $C_{\text{ant}}$  concentrations in AABW are maximum (Fig. 4a); the third period consists in the observations performed in  
292 late 2009 to 2018 when the observed variations are relatively large for  $S$  and small for  $C_{\text{ant}}$ . The mean  $C_{\text{ant}}$   
293 concentrations for each period are 7, 14 and  $13 \mu\text{mol.kg}^{-1}$ , respectively, which is consistent with the results from  
294 other studies (Table 3). The  $C_{\text{ant}}$  values for 1978-1987 can hardly be compared to other studies because very few  
295 observations were conducted in the 1980s in the SO Indian sector (Sabine et al. 1999) and because of potential  
296 biases for historical data despite their careful qualification in CARINA (Lo Monaco et al., 2010). In addition, the  
297 different methods used to estimate  $C_{\text{ant}}$  can lead to different results, especially in deep and bottom waters of the



298 SO (Vázquez-Rodríguez et al., 2009). Overall, Table 3 confirms that  $C_{\text{ant}}$  concentrations were low in the 1970s  
299 and 1980s, and reached values of the order of  $10 \mu\text{mol.kg}^{-1}$  in the 1990s, a signal not clearly captured in global  
300 data-based estimates (Gruber, 1998; Sabine et al., 2004; Waugh et al., 2006; Khatiwala et al., 2013).

301 The observations presented in this analysis, although regional, offer a complement to recent estimates of  $C_{\text{ant}}$   
302 changes evaluated between 1994 and 2007 in the top 3000 m for the global ocean (Gruber et al., 2019a). In the  
303 Enderby Basin at the horizon 2000-3000 m, the accumulation of  $C_{\text{ant}}$  from 1994 to 2007 is not uniform and ranges  
304 between 0 and  $8 \mu\text{mol.kg}^{-1}$  (Gruber et al., 2019a). At our station, in the CDW (2000-3000m) the  $C_{\text{ant}}$  concentrations  
305 were not significant in 1978-1987 ( $-2$  to  $5 \mu\text{mol.kg}^{-1}$ ) but increase to an average of  $8.7 \pm 3.0 \mu\text{mol.kg}^{-1}$  in 1998-  
306 2018 (Fig. 3a) probably due to mixing with AABW that contain more  $C_{\text{ant}}$ . Interestingly, this value is close but in  
307 the high range of the  $C_{\text{ant}}$  accumulation estimated from 1994 to 2007 in deep waters of the south Indian Ocean  
308 (Gruber et al., 2019a).

309 Not surprisingly, high  $C_{\text{ant}}$  concentrations are detected in the AABW formation regions (Table 3). The highest  $C_{\text{ant}}$   
310 concentrations in bottom waters (up to  $30 \mu\text{mol.kg}^{-1}$ ) were observed in the ventilated shelf waters in the Ross Sea  
311 (Sandrini et al., 2007). In the Adélie and Mertz Polynya regions, Shadwick et al. (2014) observed high  $C_{\text{ant}}$   
312 concentrations in the subsurface shelf waters ( $40$ – $44 \mu\text{mol.kg}^{-1}$ ) but lower values in ALBW ( $15 \mu\text{mol.kg}^{-1}$ ) when  
313 it mixed with older CDW. In WSBW, all  $C_{\text{ant}}$  concentrations estimated from observations between 1996 and 2005  
314 and with the TrOCA method (Table 3) lead to about the same values ranging between  $13$  and  $16 \mu\text{mol.kg}^{-1}$  (Lo  
315 Monaco et al., 2005b; van Heuven et al., 2011). In bottom waters formed near the Cape Darnley (CDBW), Roden  
316 et al. (2016) estimated high  $C_{\text{ant}}$  concentrations in bottom waters ( $25 \mu\text{mol.kg}^{-1}$ ) resulting from the shelf waters  
317 that contain very high  $C_{\text{ant}}$  ( $50 \mu\text{mol.kg}^{-1}$ ). The comparison with other studies confirms that far from the AABW  
318 formation sites, contemporary  $C_{\text{ant}}$  concentrations are not exceeding  $16 \mu\text{mol.kg}^{-1}$  on average. However, higher  
319  $C_{\text{ant}}$  concentrations are not unrealistic (Sandrini et al., 2007; Roden et al., 2016; this study in 2004) and likely  
320 related to ventilation and water masses variability.

321 Table 3.

## 322 5.2. $C_{\text{ant}}$ trends and variability

323 Comparison of long-term  $C_{\text{ant}}$  trends in deep and bottom waters of the SO is limited to very few regions where  
324 repeated observations are available. To our knowledge, only 3 other studies evaluated the long-term  $C_{\text{ant}}$  trends in  
325 the Southern Ocean based on more than 5 reoccupations: in the South-western Atlantic (Rios et al., 2012) and in  
326 the Weddell Gyre along the Prime meridian section (van Heuven et al., 2011, 2014). Temporal changes of  $C_T$  and  
327  $C_{\text{ant}}$  have also been investigated in other SO regions, but limited to 2 to 4 reoccupations (Murata et al., 2019;  
328 Williams et al., 2015; Pardo et al., 2017). Given the  $C_{\text{ant}}$  variability depicted at our location (Fig. 4a), different  
329 trends can be deduced from limited reoccupations. As an example, Murata et al., (2019) evaluated the change in  
330  $C_{\text{ant}}$  from data collected 17 years apart (1994–1996 and 2012–2013) along a transect around  $62^\circ\text{S}$  and found a  
331 small increase at our location ( $< 5 \mu\text{mol.kg}^{-1}$  around  $60^\circ\text{E}$ ). This result appears very sensitive to the time of the  
332 observation given that we found a minimum in  $C_{\text{ant}}$  concentrations between 2011 and 2014 (Fig. 4a) associated  
333 with a marked  $C_T$  minimum (Fig. 4b). In addition, our results show that the detection of  $C_{\text{ant}}$  trends appears very  
334 sensitive to the time period considered (Table 2). As an extreme case, the  $C_{\text{ant}}$  trend estimated for the period 1987-  
335 2004 is  $+6.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  (close to the theoretical  $C_{\text{ant}}$  trend of  $+8 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ), but it reverses to -  
336  $3.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  for the period 2004-2018.



337 The long-term  $C_T$  trend that we estimated in AABW in the eastern Enderby Basin ( $2.0 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ) is  
338 slightly faster than the  $C_T$  trends estimated in the WSBW in the Weddell Gyre:  $+1.2 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  over  
339 the period 1973-2011 and  $+1.6 \pm 1.4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  when restricted to 1996-2011 (van Heuven et al., 2014).  
340 Along the SR03 line (south of Tasmania) reoccupied in 1995, 2001, 2008 and 2011, Pardo et al (2017) evaluated  
341 a  $C_T$  trend of  $+2.4 \pm 0.2 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  in the AABW composed of ALBW and RSBW in this sector. This is  
342 higher than the  $C_T$  trends found at our location and in the Weddell Gyre, but surprisingly, this was not associated  
343 with a significant increase in  $C_{\text{ant}}$ . The  $C_T$  trend in AABW along the SR03 section was likely due to the intrusion  
344 of old and  $C_T$ -rich waters also revealed by an increase in silicate concentrations during 1995-2011 (Pardo et al.,  
345 2017). This is a clear example of decoupling between  $C_T$  and  $C_{\text{ant}}$  trends in deep and bottom waters as observed at  
346 our location in the last decade (Table 2). For  $C_{\text{ant}}$ , our 40-years trend estimate ( $1.6 \pm 0.6 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ) appears  
347 close to the trend reported by Rios et al. (2012) in the South-Western Atlantic AABW from 6 reoccupations  
348 between 1972 and 2003 ( $+1.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ). However, if we limit our result to the period 1978-2002 or  
349 1978-2004 (about the same period as in Rios et al., 2012), our trend is much larger ( $+3-4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ).  
350 At our location, the  $C_{\text{ant}}$  trend over 40 years ( $+1.6 \pm 0.6 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ) explains most of the observed  $C_T$   
351 increase ( $+2.0 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ). The residual of  $+0.4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  reflects changes in natural  
352 processes affecting the carbon content (different AABW sources, ventilation, mixing with deep waters,  
353 remineralization or carbonates dissolution). Although this is a weak signal, the natural  $C_T$  change ( $C_{T_{\text{nat}}}$ ) mirrors  
354 the observed decrease in  $O_2$  by  $-0.80 \pm 0.4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ . This  $O_2$  decrease detected in the Enderby Basin  
355 appears to be a real feature that was documented at large scale for 1960-2010 in deep SO basins (Schmidtko et al.  
356 2017), suggesting that the changes observed at  $56.5^\circ\text{S}-63^\circ\text{E}$  are related to large-scale processes, possibly due to a  
357 decrease in AABW formation (Purkey and Johnson, 2012).

### 358 5.3. Recent $C_{\text{ant}}$ stability

359 Although most studies suggest a gradual accumulation of  $C_{\text{ant}}$  in the AABW, our time-series highlights significant  
360 pluriannual changes, in particular over the last decade when  $C_{\text{ant}}$  concentrations were as low as around the year  
361 2000 (Fig. 4a) and decoupled from the increase in  $C_T$  (Fig. 4b). This result is difficult to interpret because at our  
362 location, away from AABW sources (Fig. 1), the temporal variability observed in the AABW layer can result from  
363 many remote processes occurring at the AABW formation sites (such as wind forcing, ventilation, sea-ice melting,  
364 thermodynamic, biological activity and air-sea exchanges), but internal processes during the transport of AABWs  
365 (such as organic matter remineralization, carbonate dissolution and mixing with surrounding waters) must also be  
366 taken into account. The apparent steady  $C_{\text{ant}}$  feature suggests that AABWs found at our location has stored less  
367  $C_{\text{ant}}$  in recent years. This might be linked to reduced  $\text{CO}_2$  uptake in the AABW formation regions, as recognized  
368 at large-scale in the SO from the late 1980s to 2001 (Le Quéré et al., 2007; Metzl, 2009; Lenton et al., 2012;  
369 Landschützer et al., 2015). This large-scale response in the SO during a positive trend in the Southern Annular  
370 Mode (SAM) is mainly associated to stronger winds driven by accelerating greenhouse gas emissions and  
371 stratospheric ozone depletion, leading to warming and freshening in the SO (Swart et al., 2018), change in the  
372 ventilation of the carbon-rich deep waters and reduced  $\text{CO}_2$  uptake (Lenton et al., 2009). The reconstructed  $\text{pCO}_2$   
373 fields by Landschützer et al. (2015) suggest that the reduced  $\text{CO}_2$  sink in the 1990s is identified at high latitudes  
374 in the SO (see Fig. 2a and S9 in Landschützer et al., 2015). However, as opposed to the circumpolar open ocean  
375 zone (e.g. Metzl, 2009; Takahashi et al., 2009, 2012; Munro et al., 2015; Fay et al., 2018), the long-term trend of



376 surface  $f\text{CO}_2$  and carbon uptake deduced from direct observations are not clearly identified in the seasonal ice  
377 zone (SIZ) and shelves around Antarctica, and thus in the AABW formation regions of interest to interpret our  
378 results (Laruelle et al., 2018). There, surface  $f\text{CO}_2$  data are sparse, especially before 1990, and cruises were mainly  
379 conducted in austral summer when the spatio-temporal  $f\text{CO}_2$  variability is very large and driven by multiple  
380 processes at regional or small scales, such as primary production, sea-ice formation and retreat, circulation and  
381 mixing. This leads to various estimates of the air-sea  $\text{CO}_2$  fluxes around Antarctica depending on the region and  
382 period and large uncertainty when attempting to detect long-term trends (Gregor et al., 2018).

383 In particular, in polynyas and AABW formation regions where  $f\text{CO}_2$  is low and where katabatic winds prevail,  
384 very strong instantaneous  $\text{CO}_2$  sink can occur at the local scale (up to  $-250 \text{ mmol C.m}^{-2}.\text{d}^{-1}$  in Terra Nova Bay in  
385 the Ross Sea according to DeJong and Dunbar, 2017). In the Prydz Bay region where CDBW is formed, recent  
386 studies show that surface  $f\text{CO}_2$  in austral summer vary in a very large range (150–450  $\mu\text{atm}$ ), with the lowest  $f\text{CO}_2$   
387 observed in the shelf region generating very strong local  $\text{CO}_2$  sink ( $-221 \text{ mmol C.m}^{-2}.\text{d}^{-1}$  according to Roden et al.  
388 2016). The carbon uptake was particularly enhanced near Cape Darnley and coincided with the highest  $C_{\text{ant}}$   
389 concentrations that Roden et al. (2016) estimated in the dense shelf waters that subduct to form AABW. In the  
390 Prydz Bay coastal region, surface  $f\text{CO}_2$  values in 1993–1995 were as low as 100  $\mu\text{atm}$  (Gibson and Trull, 1999)  
391 leading to a strong local  $\text{CO}_2$  uptake of  $-30 \text{ mmol C.m}^{-2}.\text{d}^{-1}$  in summer. In addition, Roden et al. (2013) found a  
392 large  $C_T$  increase over 16 years ( $+34 \mu\text{mol.kg}^{-1}$ ) in the Prydz Bay, which is much higher than the anthropogenic  
393 signal alone ( $+12 \mu\text{mol.kg}^{-1}$ ) and likely explained by changes in primary production that would have been stronger  
394 in 1994. To our knowledge, this is the only direct observation of decadal  $C_T$  change in surface waters in a region  
395 of AABW formation (here the Prydz Bay) and it highlights the difficulty not only to evaluate the  $C_T$  and  $C_{\text{ant}}$  long-  
396 term trends in these regions but also to separate natural and anthropogenic signals when this water reaches the  
397 deep ocean. We attempted to detect long-term changes in  $\text{CO}_2$  uptake in this region using the qualified  $f\text{CO}_2$  data  
398 available in the SOCAT database (Bakker et al., 2016), but our estimates (not shown) were highly uncertain due  
399 to very large spatial and temporal variability. To conclude, all previous studies conducted near or in AABW  
400 formation sites clearly reveal that these regions are potentially strong carbon sinks, but how the sink changed over  
401 the last decades is not yet evaluated, and thus we are not able to certify that the recent  $C_{\text{ant}}$  stability that we observed  
402 in the AABW at our location is directly linked to the weakening of the carbon sink that was recognized at large-  
403 scale in the SO from the 1980s to mid-2000s (Le Quéré et al., 2007; Landschützer et al., 2015).

404 Changes in the accumulation of  $C_{\text{ant}}$  in AABW could also be directly related to changes in physical processes  
405 occurring in AABW formation region. Decadal decreasing of sea-ice production and melting of sea-ice have been  
406 documented in several regions including Cape Darnley polynyas (Tamura et al., 2016; Williams et al., 2016). The  
407 consequent changes in Antarctic surface waters properties are transmitted into the deep ocean, notably the well-  
408 recognized freshening of AABWs over the last decades (Rintoul, 2007). The warming of bottom waters was also  
409 documented in the Enderby basin (Coudrey et al., 2013) as well as at a larger scale in all deep SO basins (Purkey  
410 and Johnson, 2010; Desbruyères et al., 2016). Associated to a decrease in AABW formation in the 1990s (Purkey  
411 and Johnson, 2012), these physical changes could explain the recent stability of  $C_{\text{ant}}$  concentrations in AABW  
412 observed at our location. As AABWs from different sources spread and mix with  $C_T$ -rich deep waters before  
413 reaching our location (Fig. 1), less AABW formation and export would result in an increase in  $C_T$  (increase in  
414  $C_{\text{Tnat}}$ ) not associated with an increase in  $C_{\text{ant}}$ , and a decrease in  $\text{O}_2$  (as observed in recent years in Fig. 4a,b,c).  
415 Finally, it is also possible that the AABW observed in recent years at our location is the result of a larger



416 contribution of older RSBW and/or ALBW that have lower  $C_{\text{ant}}$  and  $O_2$  concentrations compared to CDBW formed  
417 at Cape Darnley and Prydz Bay.

## 418 6. Conclusion

419 The distribution and evolution of  $C_{\text{ant}}$  in the bottom layer of the SO are related to complex interactions between  
420 climatic forcing, air-sea  $CO_2$  exchange at formation sites, as well as biological and physical processes during  
421 AABWs circulation. The dataset that we collected regularly in the Enderby basin over the last 20 years (1998-  
422 2018) in the frame of the OISO project, together with historical observations obtained in 1978, 1985 and 1987  
423 (GEOSECS and INDIGO cruises), allows the investigation of  $C_{\text{ant}}$  changes in AABW over 40 years in this region.  
424 Our results suggest that the accumulation of  $C_{\text{ant}}$  explains most of (but not all) the observed increase in  $C_T$ . We  
425 also detected a decrease in  $O_2$  that is consistent with the large-scale signal reported by Schmidtko et al. (2017),  
426 possibly due to a decrease in AABW formation (Purkey and Johnson, 2012). Our data further indicate rapid  
427 anomalies in some periods suggesting that for decadal to long-term estimates care have to be taken when analyzing  
428 the change in  $C_{\text{ant}}$  from data sets collected 10 or 20 years apart (e.g. Williams et al., 2015; Murata et al., 2019).  
429 Our results also show different  $C_{\text{ant}}$  trends on short periods, with a maximum increase of  $6.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$   
430 between 1987 and 2004 and an apparent stability in the last 20 years (despite an increase in  $C_T$ ). This suggests that  
431 AABWs have stored less  $C_{\text{ant}}$  in the last decade, but our understanding of the processes that explain this signal is  
432 not clear. This might be the result of the reduced  $CO_2$  uptake in the SO in the 1990s (Le Quéré et al., 2007;  
433 Landschützer et al., 2015), but this is not yet verified from direct  $C_T$  or  $fCO_2$  observations in AABW formation  
434 regions due to the lack of winter data and very large variability during summer. This calls for more data collection  
435 and investigations in these regions. The apparent stability of  $C_{\text{ant}}$  in AABW since 1998 could also be directly linked  
436 to a decrease in AABW formation in the 1990s (Purkey and Johnson, 2012) or a change in the contributions of  
437 AABWs from different sources, especially in the Prydz Bay region (Williams et al., 2016). In these scenarios, an  
438 increased contribution of  $C_T$ -rich and  $O_2$ -poor older CDW would also explain the decoupling between  $C_{\text{ant}}$  and  $C_T$   
439 (increase in  $C_{T_{\text{nat}}}$ ) and decrease in  $O_2$  concentrations observed in recent years. The decoupling between  $C_{\text{ant}}$  and  
440  $C_T$  is not a unique feature, as it was also reported along the SR03 section between Tasmania and Antarctica, most  
441 probably due to advection of  $C_T$ -rich waters (Pardo et al., 2017). This highlights the importance of the ocean  
442 circulation in influencing the temporal  $C_T$  and  $C_{\text{ant}}$  inventories changes (De Vries et al. 2017) and the need to better  
443 separate anthropogenic and natural variability based on time-series observations.  
444 The evaluation and understanding of decadal  $C_{\text{ant}}$  changes in deep and bottom ocean waters are still challenging,  
445 as the  $C_{\text{ant}}$  concentrations remain low compared to  $C_T$  measurements accuracy (at best  $\pm 2 \mu\text{mol.kg}^{-1}$ , Bockmon  
446 and Dickson, 2015) and uncertainties of data-based methods ( $\pm 6 \mu\text{mol.kg}^{-1}$ ). Long-term repeated and qualified  
447 observations (at least 30 years) are needed to accurately detect and separate the anthropogenic signal from the  
448 internal ocean variability; we thus only start to document these trends that should now help to identify  
449 shortcomings in models regarding the carbon storage in the deep SO (e.g., Frölicher et al., 2014). As changes in  
450 the SO (including warming, freshening, oxygenation/deoxygenation,  $CO_2$  and acidification) are expected to  
451 accelerate in the future in response to anthropogenic forcing and climate change (e.g. Hauck et al., 2015; Heuzé et  
452 al., 2014; Ito et al., 2015, Yamamoto et al., 2015), it is important to maintain time-series observations to  
453 complement the GO-SHIP strategy, and to occupy more regularly other sectors of the SO (Rintoul et al., 2012). In



454 this context, we hope to maintain our observations in the Southern Indian Ocean in the next decade, and with  
455 ongoing synthetic products activities such as GLODAPv2 (Olsen et al., 2016; 2019), SOCAT (Bakker et al., 2016)  
456 and more recently the SOCCOM project (Williams et al., 2018), to offer a solid database to validate ocean  
457 biogeochemical models and coupled climate/carbon models (Russell et al. 2018), and ultimately reduce  
458 uncertainties in future climate projections.

#### 459 **Data availability**

460 GEOSECS, INDIGO and OISO 1998-2011 data are publicly available at the Ocean Carbon Data System (OCADS;  
461 [https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2\\_2019](https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2_2019)). OISO original data stations are available at:  
462 [www.nodc.noaa.gov/ocads/oceans/RepeatSections/clivar\\_oiso.html](https://www.nodc.noaa.gov/ocads/oceans/RepeatSections/clivar_oiso.html). OISO 2012-2018 will be available in  
463 GLODAPv2\_2021.

#### 464 **Author contributions**

465 LM, CLM, NM, JF and CM performed the sampling and carried out the measurements of the OISO data. LM  
466 prepared the manuscript with contributions from CLM and NM.

#### 467 **Competing interests**

468 The authors declare that they have no conflict of interest.

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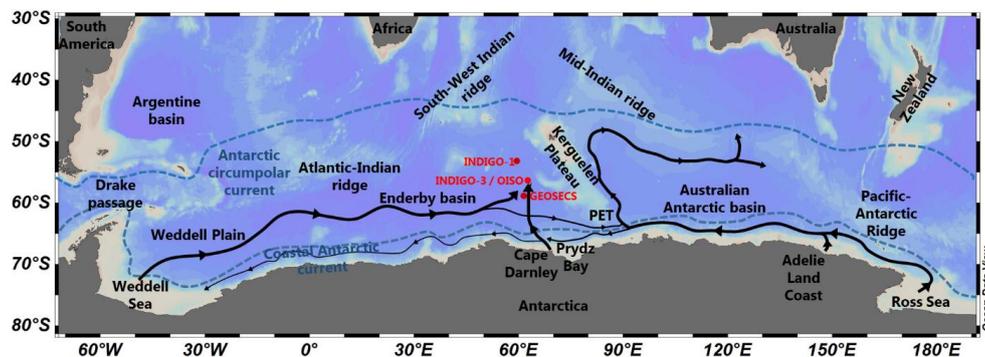
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802 **Table 1. List of the cruises used in this study.**

Cruise	Station	Location	Year	Month
GEOSECS	430	61.0°E / 60.0°S	1978	February
INDIGO-1	14	58.9°E / 53.0°S	1985	March
INDIGO-3	75	63.2°E / 56.5°S	1987	January
OISO-01	11	63.0°E / 56.5°S	1998	February
OISO-03	11	63.0°E / 56.5°S	1998	December
OISO-05	11	63.0°E / 56.5°S	2000	August
OISO-06	11	63.0°E / 56.5°S	2001	January
OISO-08	11	63.0°E / 56.5°S	2002	January
OISO-11	11	63.0°E / 56.5°S	2004	January
OISO-18	11	63.0°E / 56.5°S	2009	December
OISO-19	11	63.0°E / 56.5°S	2011	January
OISO-21	11	63.0°E / 56.5°S	2012	February
OISO-23	11	63.0°E / 56.5°S	2014	January
OISO-26	11	63.0°E / 56.5°S	2016	October
OISO-27	11	63.0°E / 56.5°S	2017	January
OISO-28	11	63.0°E / 56.5°S	2018	January

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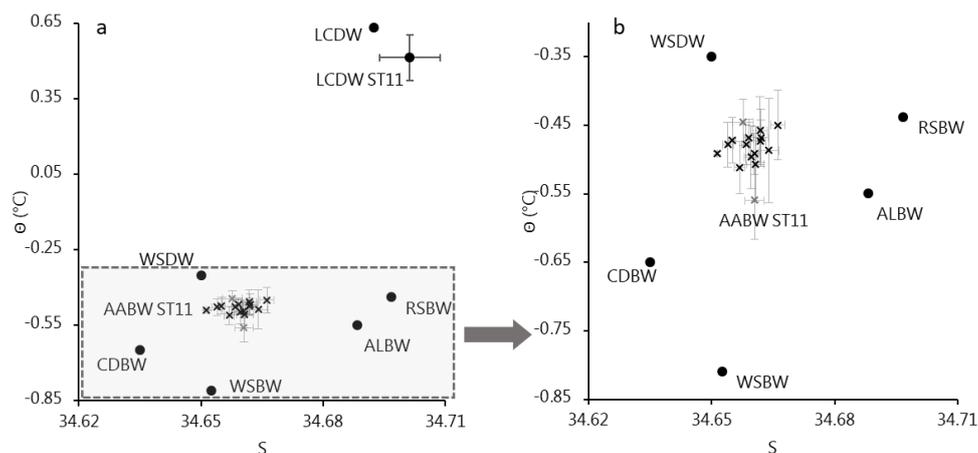


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 808 **Figure 1. The AABWs circulation from the literature (Fukamachi et al., 2010; Orsi et al., 1999) and this study, with**  
 809 **geographic indications (black text), SO currents (blue text and dash lines for the approximative positions) and stations**  
 810 **considered in this study (red text and dots). PET: Princess Elizabeth Trough. Figure produced with ODV (Schlitzer et**  
 811 **al., 2019).**

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816 **Figure 2. (a) Full  $\Theta$ -S diagram of studied water masses and (b) zoomed on bottom waters. Values are from literature**  
817 **for the WSBW (Fukamachi et al., 2010; van Heuven, 2013; Pardo et al., 2014; Robertson et al., 2002), the WSDW**  
818 **(Carmack and Foster, 1975; Fahrback et al., 1994; van Heuven, 2013; Robertson et al., 2002), the RSBW (Fukamachi**  
819 **et al., 2010; Gordon et al., 2015; Johnson, 2008; Pardo et al., 2014), the ALBW (Fukamachi et al., 2010; Johnson, 2008;**  
820 **Pardo et al., 2014), the CDBW (Ohshima et al., 2013) and the LCDW (Lo Monaco et al., 2005a; Pardo et al., 2014; Smith**  
821 **and Treguer, 1994), and from the OISO-ST11 dataset for the OISO-ST11 AABW and OISO-ST11 LCDW. Error bars**  
822 **are calculated from the individual annual averaged values for the OISO-ST11 AABW and from all data for the OISO-**  
823 **ST11 LCDW. For the OISO-ST11 AABW, the grey cross are the GEOSECS (lowest  $\Theta$ ) and INDIGO-1 (highest  $\Theta$ )**  
824 **values.**  
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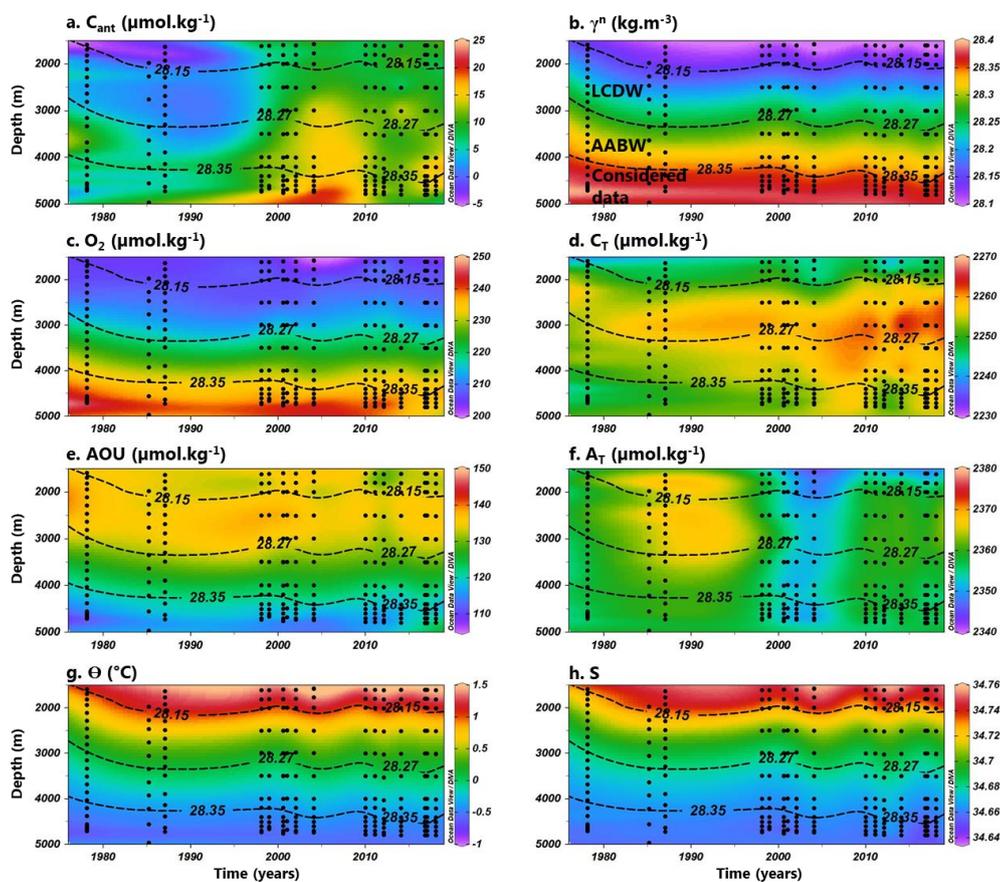
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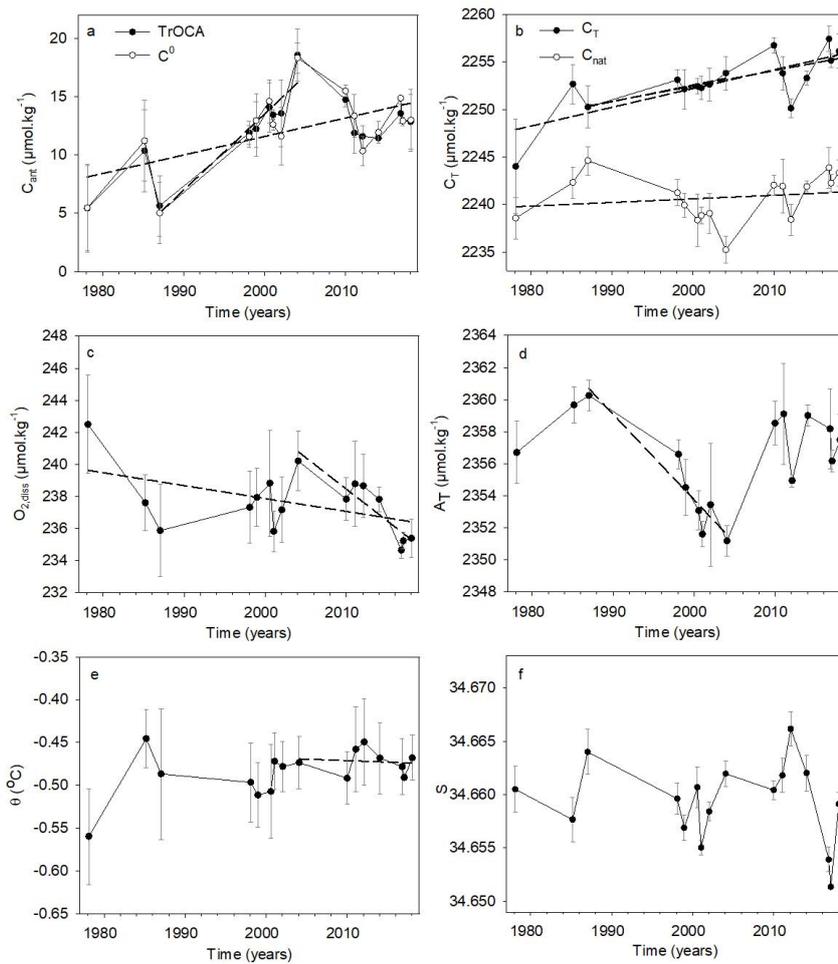
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832 Figure 3. Hovmöller section of the carbon related properties parameters ( $C_{\text{ant}}$  via TrOCA,  $O_2$ , AOU,  $C_T$  and  $A_T$ ) and  
833 hydrological properties ( $\Theta$ ,  $S$  and  $\gamma^n$ ) of the dataset presented in Table 1 from 1978 to 2018 and from 1500 m to the  
834 bottom. Data points are represented by the black dots. The dash isolines represent the water masses separation by  $\gamma^n$   
835 detailed on the  $\gamma^n$  plot. Figure produced with ODV (Schlitzer et al., 2019).

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838 **Figure 4.** Interannual variability (full lines) and significant trends (at 95%, see Table 2; dotted lines) for the 40 years of  
 839 observation of the OISO-ST11 AABW properties, including (a)  $C_{ant}$  by the TrOCA and the  $C^o$  method, (b)  $C_T$  and  $C_{nat}$ ,  
 840 (c)  $O_2$ , (d)  $A_T$ , (e)  $\theta$  and (f)  $S$ .

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851 **Table 2: Trends (per decade) of observed and calculated properties in the AABW layer estimated over different periods**  
 852 **(in bold: significant trends at 95% confidence level).**

Period	S	Θ °C	Silicate μmol.kg <sup>-1</sup>	Nitrate μmol.kg <sup>-1</sup>	O <sub>2</sub> μmol.kg <sup>-1</sup>	AOU μmol.kg <sup>-1</sup>	A <sub>T</sub> μmol.kg <sup>-1</sup>	C <sub>T</sub> μmol.kg <sup>-1</sup>	C <sub>ant</sub> TrOCA μmol.kg <sup>-1</sup>
<b>1978-2018</b>	-0.001 ± 0.001	0.01 ± 0.01	-1.2 ± 0.9	0.2 ± 0.2	<b>-0.8 ± 0.4</b>	<b>0.7 ± 0.0</b>	-0.1 ± 0.1	<b>2.0 ± 0.5</b>	<b>1.6 ± 0.6</b>
<b>1987-2018</b>	-0.001 ± 0.001	0.01 ± 0.01	-1.9 ± 1.4	0.3 ± 0.4	-0.3 ± 0.5	0.2 ± 0.5	0.6 ± 0.1	<b>1.6 ± 0.5</b>	1.1 ± 0.8
<b>1987-2004</b>	-0.003 ± 0.002	0.01 ± 0.01	<b>-6.5 ± 1.8</b>	0.9 ± 0.9	1.7 ± 1.0	-1.7 ± 1.0	<b>-5.3 ± 0.1</b>	<b>1.8 ± 0.4</b>	<b>6.5 ± 1.0</b>
<b>2004-2018</b>	-0.006 ± 0.003	<b>0.02 ± 0.01</b>	-1.8 ± 4.5	-0.5 ± 1.0	<b>-3.9 ± 0.7</b>	<b>4.0 ± 0.8</b>	3.4 ± 0.2	1.7 ± 1.9	-3.5 ± 1.5

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856 **Table 3. Compilation of C<sub>ant</sub> sequestration investigations in the AABWs ( $\gamma^n \geq 28.25 \text{ kg.m}^{-3}$ ) using the TrOCA method.**  
 857 **The C<sub>ant</sub> estimation of Pardo et al. (2014) is calculated using theoretical AABW mean composition (with 3% of ALBW)**  
 858 **and the carbon data from the GLODAPv1 and CARINA databases. Sandrini et al. (2007) values has been measured at**  
 859 **the bottom in the Ross Sea and correspond to recently sunk HSSW. The mean values published by Roden et al. (2016)**  
 860 **for the AABWs present WSDW characteristics but can be a mix of CDBW and CDW.**

Source	Location	Water masses considered	Year	C <sub>ant</sub> μmol.kg <sup>-1</sup>
Pardo et al. (2014) Fig. 5	Averaged AABW composition	WSBW-RSBW- ALBW	1994	12
Lo Monaco et al. (2005b) Fig. 4b	WOCE line I6 (30°E; 50°-70°S)	WSBW CDBW	1996	15 20
Sandrini et al. (2007) Fig. 4a	Ross Sea	HSSW (previous RSBW)	2002/2003	Max. of 30
Shadwick et al. (2014) Table 2	Mertz polynya and Adelie depression	ALBW	2007/2008	15
Roden et al. (2016) Table 2	South Indian ocean (30°-80°E; 60°-69°S)	WSDW-CDW- CDBW	2006	25
van Heuven et al. (2011) Fig.13	Weddell gyre (0°E; 55°-71°S)	WSBW	2005	16
			1978-1987	7 ± 3
			1987-1998	9 ± 4
This study	Enderby basin (56.5°S-63°E)	WSDW-CDBW- RSBW-ALBW	1987-2004	13 ± 4
			1998-2004	14 ± 2
			2010-2018	13 ± 1
			<b>1978-2018</b>	<b>12 ± 3</b>

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