



# Variability and stability of anthropogenic CO2 in Antarctic

# 2 Bottom Waters observed in the Indian sector of the Southern

# 3 Ocean, 1978-2018

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#### 8 Abstract

- 9 Antarctic bottom waters (AABWs) are known as a long term sink for anthropogenic CO<sub>2</sub> (C<sub>ant</sub>) but is hardly
- 10 quantified because of the scarcity of the observations, specifically at an interannual scale. We present in this
- 11 manuscript an original dataset combining 40 years of carbonate system observations in the Indian sector of the
- 12 Southern Ocean (Enderby Basin) to evaluate and interpret the interannual variability of Cant in the AABW. This
- 13 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
- 15 1987).
- 16 At this location the main sources of AABW sampled is the fresh and younger Cape Darnley bottom water (CDBW)
- 17 and the Weddell Sea deep water (WSDW). Our calculations reveal that Cant concentrations increased significantly
- 18 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
- 19 estimates in other SO basins, with the exception of bottom waters close to their formation sites where Cant
- 20 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  $C_T$  is mainly due to the
- 22 uptake of anthropogenic CO<sub>2</sub> in the different formation regions. This is however modulated by significant
- 23 interannual to pluriannual variability associated with variations in hydrological (Θ, S) and biogeochemical (C<sub>T</sub>,
- $A_T$ ,  $O_2$ ) properties. A surprising result is the apparent stability of  $C_{ant}$  concentrations in recent years despite the
- 25 increase in C<sub>T</sub> and the gradual acceleration of atmospheric CO<sub>2</sub>.
- 26 The Cant sequestration by AABWs is more variable than expected and depends on a complex combination of
- 27 physical, chemical and biological processes at the formation sites and during the transit of the different AABWs.
- 28 The interannual variability at play in AABW needs to be carefully considered on the extrapolated estimation of
- 29 C<sub>ant</sub> sequestration based on sparse observations over several years.

## 1 Introduction

30 31

- 32 CO<sub>2</sub> atmospheric concentration has been increasing since the start of the industrialization (Keeling and Whorf,
- 33 2000). This increase leads to an ocean uptake of about a quarter of C<sub>ant</sub> emissions (Le Quéré et al., 2018; Gruber
- 34 et al., 2019a). It is widely acknowledged that the Southern Ocean (SO) is responsible for 40 % of the Cant ocean
- 35 sequestration (Gruber et al., 2009; Khatiwala et al., 2009; Matear, 2001; McNeil et al., 2003; Orr et al., 2001).
- 36 Ocean Cant uptake and sequestration have the benefit to limit the atmospheric CO2 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<sub>ant</sub> sequestration and its variability is of major importance to predict future atmospheric CO<sub>2</sub> concentrations, impact 38 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 Cant in seawater cannot be measured directly and the evaluation of the relatively small Cant signal from the total 42 inorganic dissolved carbon (CT; 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 Cant concentrations in the oceans. The 'historical' 44 back calculation method based on C<sub>T</sub> measurement and preformed inorganic carbon estimate (C<sup>0</sup>) was 45 independently published by Brewer (1978) and Chen and Millero (1979). This method has been often applied at regional and basin scale (Chen, 1982, 1993; Goyet et al., 1998; Körtzinger et al., 1998, 1999; Poisson and Chen, 46 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 in various regions including the SO (e.g. Lo Monaco et al., 2005b; Sandrini et al., 2007; Pardo et al., 2014; Roden 49 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 Cant concentrations, especially at high latitudes and more particularly in deep and bottom waters (Lo Monaco et al., 2005b; Vázquez-Rodríguez et al., 2009; Pardo et al., 2014). Thus, there 52 53 is a need to better explore the C<sub>T</sub> and C<sub>ant</sub> 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 CO2 and heat regulation as they play a major role in the meridional overturning circulation (Johnson et al., 2008; Marshall and Speer, 2012) . 56 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 Cant 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 constantly pushed away by the wind, leading to cold and salty surface waters in contact with the atmosphere. The 66 variable conditions of wind, ice production, surface water cooling and continental slope shape encountered around 67 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 Cant has been controversial since one can believe that the ice coverage limits 71 the invasion of Cant 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 Cant signals in AABW formation regions, likely due to the 73 uptake of CO<sub>2</sub> 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 CO2 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





- al., 2011), the amount of C<sub>ant</sub> stored in the AABWs may be very variable, which could bias the estimates of C<sub>ant</sub>
- 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<sub>ant</sub> 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<sub>ant</sub> and C<sub>T</sub> 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)
- 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
- 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
- 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





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

#### 2.3 AABW definition

- 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 kg.m<sup>-3</sup> 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<sub>ant</sub> concentrations during AABW formation (Shadwick et al., 2013). The Θ-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  $60^{\circ}$  S (-0.56
- °C), probably because it experienced less mixing with CDW compared to the warmer type of AABW observed at
- the INDIGO-1 station at 53° S (-0.44 °C). For the other cruises and years, θ in AABW ranges from -0.51 to -0.45
   °C with no clear indication on the specific AABW origin. The S range observed in the bottom waters at OISO-
- ST11 (34.65-34.67), illustrates either changes in mixing with various AABW sources or temporal variations at the
- formation site. Given the knowledge of deep and bottom waters circulation and characteristics (Fig.s 1 and 2) and
- the significant C<sub>ant</sub> concentrations that we estimated at depth (Fig. 3), the main contribution at our location is likely
- the younger and colder CDBW for which relatively high Cant 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
- 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 Cant 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
- 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
- and INDIGO were qualified in GLODAP-v1 (Key et al., 2004) and previously used for the first C<sub>ant</sub> estimates in





- 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<sub>T</sub> 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<sub>ant</sub> 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,
- 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. Θ (in °C) and S (no unit) measurements have an
- 161 accuracy of 0.01 of their respective units. C<sub>T</sub> and A<sub>T</sub> were sampled in 500 mL glass bottles and poisoned with 100
- 162 µL of HgCl<sub>2</sub> saturated solution to halt biological activity. Discrete C<sub>T</sub> and A<sub>T</sub> samples were analyzed onboard by
- 163 potentiometric titration derived from the method developed by Edmond (1970) using a closed cell. The accuracy
- for  $C_T$  and  $A_T$  varies from 1 to 3.5  $\mu$ mol.kg<sup>-1</sup> (depending on the cruise) and is determined by sample duplicates in
- surface, at 1000 m and bottom waters. All measurements were calibrated with Certified Reference Materials (CRMs) provided by A.G. Dickson laboratory (Scripps Institute of Oceanography). O<sub>2</sub> was determined by an
- oxygen sensor fixed on the rosette. These values were adjusted using measurements obtained by Winkler titrations
- using a potentiometric titration system (at least 12 measurements for each profile). Thiosulphate solution used in
- Winkler titration was calibrated using iodate standard solution to provide the standard O<sub>2</sub> accuracy of 2 μmol.kg
- 170 <sup>1</sup>. Nitrate (NO<sub>3</sub>) and Silicate (Si) were measured onboard or offshore with an automatic colorimetric Technicon
- analyser following the methods described by Tréguer and Le Corre (1975) until 2008, and the revised protocol
- described by Aminot and Kérouel (2007) since 2009. Based on replicate measurements for deep samples we
- estimate an error of about 0.3 % for both nutrients. NO<sub>3</sub> concentrations are not available for all the cruises used
- in this analysis. The mean NO<sub>3</sub> concentrations in the AABW at OISO-ST11 is  $32.8 \pm 1.2 \,\mu\text{mol.kg}^{-1}$  while the
- 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 μmol.kg<sup>-1</sup>. The lack of NO<sub>3</sub> for few cruises has been palliated by considering a standard value of 33 μmol.kg<sup>-1</sup>
- 177 with a limited impact on C<sub>ant</sub> determined by the C° method (from 0.1 μmol.kg<sup>-1</sup> to 1.7 μmol.kg<sup>-1</sup> on the mean
- 178 annual values).

# 179 3.3 Cant 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
- a combination of O<sub>2</sub>, C<sub>T</sub> and A<sub>T</sub>, following:

183 
$$TrOCA = O_2 + a\left(C_T - \frac{1}{2}A_T\right),$$
 (1)

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, Cant can be directly calculated from the difference between
- 187 TrOCA and its pre-industrial value TrOCA°:

$$C_{ant} = \frac{TrocA - TrocA^0}{a}, \tag{2}$$

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

190 
$$TrOCA^0 = e^{\left[b - (c).\theta - \frac{d}{A_T^2}\right]},$$
 (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<sub>ant</sub> is:

195

$$C_{ant} = \frac{o_{2} + 1,279 \left(c_{T} - \frac{1}{2}A_{T}\right) - e^{\left[7,511 - (1,087.10^{-2}).\Theta - \frac{7,81.10^{5}}{A_{T}^{2}}\right]}}{1,279},$$
(4)

The consideration of the errors on the different parameters involved in the TrOCA method results in an uncertainty

 $197 \qquad \text{of } \pm 6.25 \; \mu\text{mol.kg}^{\text{-}1} \; (\text{mostly due to the parameter a, leading to } \pm 3.31 \; \mu\text{mol.kg}^{\text{-}1}). \; \text{As this error is relatively large}$ 

compared to the expected Cant concentrations in deep and bottom SO waters (Pardo et al., 2014) we will compare

the TrOCA results using another indirect method to interpret Cant changes over 40 years.

## 200 3.4 Cant calculation using the preformed inorganic carbon method

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

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

206 Cbio (Eq. 6) depends on carbonate dissolution and organic matter remineralization, taking account of the corrected

207 Redfield ratio from Kortzinger et al. (2001):

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

- 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:

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

- 213 PO (Eq. 8) has been defined by Broecker (1974) and depends on the equilibrium of O<sub>2</sub> with phosphate (PO<sub>4</sub>). When
- PO<sub>4</sub> data are not available, nitrate (NO<sub>3</sub>) can be used instead (Anderson and Sarmiento, 1994):

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

- 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
- and Krause, 1980) and after only impacted by the biological activity (Weiss, 1970). The correction of  $O_{2,sat}$  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
- al., 2005a), and a mean value for  $O_2$  undersaturation in ice-covered surface waters  $\alpha$ =12 % (Anderson et al., 1991)
- according to Eq. 9:





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

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

$$224 C_{0,PI} = C_{0,obs} + \left[C_T - C_{bio} - C_{0,obs}\right]_{p_{FF}}, (10)$$

- 225 Where the reference water term has been computed using an optimum multiparametric (OMP) model and defined
- 226 as 51 µmol.kg<sup>-1</sup> from North Atlantic deep water (Lo Monaco et al., 2005a) and C<sub>0.obs</sub> has been computed similarly
- 227 as A<sub>T</sub><sup>0</sup> (Eq. 11):

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

- 229 For more details in the  $C^{\circ}$  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
- their evolution over the last 40 years are displayed in Fig. 3. The LCDW layer ( $\gamma^n = 28.15 28.27 \text{ kg.m}^{-3}$ ) is
- 234 characterized by maximum AOU values (Fig. 3c), maximum C<sub>T</sub> concentrations (Fig. 3d) and minimum C<sub>ant</sub>
- 235 concentrations (Fig. 3a). Cant concentrations were not significant in the LCDW until the end of the 1990s (<6
- μmol.kg<sup>-1</sup>), then our data show a sudden increase in C<sub>ant</sub> between January and December 1998, followed by
- $237 \qquad \text{relatively constant $C_{ant}$ concentrations (10\pm3\ \mu\text{mol.kg}^{-1})$. In the core of AABW ($\gamma^n > 28.35\ kg.m^{-3})$, well identified}$
- by low  $\Theta$ , low S, high  $O_2$  and low AOU,  $C_{ant}$  concentrations are higher than in the overlying deep waters (Fig. 3a)
- and increased from 5±4  $\mu$ mol.kg<sup>-1</sup> in 1978, 7±4  $\mu$ mol.kg<sup>-1</sup> in the mid-1980s to 13±2  $\mu$ mol.kg<sup>-1</sup> at the end of the
- 240 1990s and up to  $19\pm2~\mu mol.kg^{-1}$  in 2004 (Fig. 4a). Figure 4a also shows a very good agreement between the TrOCA
- 241 method and the C<sup>0</sup> method for both the magnitude and variability of C<sub>ant</sub> in the core of AABW. Our results show
- a mean  $C_{ant}$  trend in AABW of +1.6  $\mu$ mol.kg<sup>-1</sup>.decade<sup>-1</sup> over the full period and a maximum trend of the order of
- 243 +6.5 \text{ \text{\pmol.kg}^{-1}.decade}^{-1} \text{ over } 1987-2004 \text{ (Table 2)}. These trends are lower than the theoretical trend expected from
- the increase in atmospheric CO<sub>2</sub>. Indeed, assuming that the surface ocean fCO<sub>2</sub> follows the atmospheric growth
- rate (+1.8 µatm.year<sup>-1</sup> over 1978-2018), the theoretical C<sub>ant</sub> trend at the AABW formation sites would be of the
- order of +8 µmol.kg<sup>-1</sup>.decade<sup>-1</sup>. The observed slow C<sub>ant</sub> 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<sub>ant</sub> (Fig. 3).
- 248 Figure 3.
- 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
- between  $C_T$  and  $C_{ant}$ ). Over the full period,  $C_T$  increased by  $2.0\pm0.5~\mu mol.kg^{-1}.decade^{-1}$ , mostly due to the
- accumulation of C<sub>ant</sub> (Table 2). Our data also show a significant decrease in O<sub>2</sub> concentrations by 0.8±0.4 μmol.kg<sup>-1</sup>
- 253 1.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<sub>2</sub> loss in the global ocean. In the deep Indian SO sector, these
- authors found a trend approaching -1 μmol.kg<sup>-1</sup>.decade<sup>-1</sup> over 50 years (1960-2010), which is consistent with our
- data. We did not detect any significant trend in  $A_T$ ,  $\Theta$  and S over the full period, but on shorter periods our data
- show a significant decrease in A<sub>T</sub> 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<sub>Tnat</sub> (i.e. for this period the increase in C<sub>T</sub> was lower than the





O2 (increase in AOU) and a decrease in Cant (Table 2). These recent trends were associated with a small increase 261 262 in  $\theta$  (Fig. 4e, Table 2), but no significant trend in S (Fig. 4f). The increase in  $C_{Tnat}$  is thus unlikely originating 263 solely from increased mixing with LCDW during bottom waters transport. Enhanced organic matter remineralization is also unlikely since nitrate did not show any significant trend (Table 2). 264 265 Table 2. 266 Figure 4. Importantly, our data show substantial interannual variations in AABW properties, which could significantly 267 impact the trends estimated from limited reoccupations (e.g. Williams et al., 2015; Pardo et al., 2017; Murata et 268 al., 2019). For example, we found relatively higher Cant concentrations in 1985 (10 µmol.kg<sup>-1</sup>) compared to 1978 269 270 and 1987 (5 µmol.kg<sup>-1</sup>). 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<sub>T</sub> and O<sub>2</sub>, and that could explain the relatively low C<sub>ant</sub> 273 concentrations observed over this period. Indeed, the S maximum observed in 2012 (correlated to higher  $\theta$ ) is 274 associated with a marked C<sub>T</sub> minimum (surprisingly almost as low as in 1987), as well as low A<sub>T</sub> (hence low C<sub>Tnat</sub>), 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 Cant 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<sub>T</sub> and a rapid decrease in O<sub>2</sub> 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.decade<sup>-1</sup> from 2007 to 2016 283 (against -0.002.decade<sup>-1</sup> between 1994 and 2007), suggesting an acceleration of the AABW freshening in recent 284 years. However, they also reported a warming by +0.06 °C.decade<sup>-1</sup>, while we observed cooler temperature in

accumulation of Cant). This event is followed by an increase in C<sub>Tnat</sub> since 2004 associated to a rapid decrease in

## 5. Discussion

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# 5.1. Cant concentrations

288 In order to compare our Cant estimates with other studies, we separated the 40-years time-series into 3 periods: the first period (1978-1987) corresponds to historical data when C<sub>ant</sub> is expected to be low; the second period (1998-2004) starts when the first OISO cruise was conducted (and using CRMs for A<sub>T</sub>, C<sub>T</sub> measurements) and lasts when 290 Cant 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 Cant. The mean Cant concentrations for each period are 7, 14 and 13 µmol.kg<sup>-1</sup>, respectively, which is consistent with the results from other studies (Table 3). The Cant values for 1978-1987 can hardly be compared to other studies because very few observations were conducted in the 1980s in the SO Indian sector (Sabine et al. 1999) and because of potential biases for historical data despite their careful qualification in CARINA (Lo Monaco et al., 2010). In addition, the 296 different methods used to estimate Cant can lead to different results, especially in deep and bottom waters of the

2016-2018. This suggests that we sampled a different mixture of AABWs.





298 SO (Vázquez-Rodríguez et al., 2009). Overall, Table 3 confirms that C<sub>ant</sub> concentrations were low in the 1970s and 1980s, and reached values of the order of  $10 \ \mu mol.kg^{-1}$  in the 1990s, a signal not clearly captured in global 299 300 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 Cant changes evaluated between 1994 and 2007 in the top 3000 m for the global ocean (Gruber et al., 2019a). In the 302 Enderby Basin at the horizon 2000-3000 m, the accumulation of  $C_{\text{ant}}$  from 1994 to 2007 is not uniform and ranges 303 304 between 0 and 8 µmol.kg<sup>-1</sup> (Gruber et al., 2019a). At our station, in the CDW (2000-3000m) the C<sub>ant</sub> concentrations 305 were not significant in 1978-1987 (-2 to 5 μmol.kg<sup>-1</sup>) but increase to an average of 8.7±3.0 μmol.kg<sup>-1</sup> in 1998-306 2018 (Fig. 3a) probably due to mixing with AABW that contain more Cant. Interestingly, this value is close but in the high range of the Cant accumulation estimated from 1994 to 2007 in deep waters of the south Indian Ocean 307 308 (Gruber et al., 2019a). 309 Not surprisingly, high Cant concentrations are detected in the AABW formation regions (Table 3). The highest Cant concentrations in bottom waters (up to 30 µmol.kg<sup>-1</sup>) were observed in the ventilated shelf waters in the Ross Sea 310 311 (Sandrini et al., 2007). In the Adélie and Mertz Polynya regions, Shadwick et al. (2014) observed high Cant concentrations in the subsurface shelf waters (40-44 µmol.kg<sup>-1</sup>) but lower values in ALBW (15 µmol.kg<sup>-1</sup>) when 312 it mixed with older CDW. In WSBW, all Cant concentrations estimated from observations between 1996 and 2005 313 314 and with the TrOCA method (Table 3) lead to about the same values ranging between 13 and 16 µmol.kg<sup>-1</sup> (Lo 315 Monaco et al., 2005b; van Heuven et al., 2011). In bottom waters formed near the Cape Darnley (CDBW), Roden et al. (2016) estimated high C<sub>ant</sub> concentrations in bottom waters (25 µmol.kg<sup>-1</sup>) resulting from the shelf waters 316 that contain very high C<sub>ant</sub> (50 µmol.kg<sup>-1</sup>). The comparison with other studies confirms that far from the AABW 317 318 formation sites, contemporary Cant concentrations are not exceeding 16 µmol.kg<sup>-1</sup> on average. However, higher

Cant concentrations are not unrealistic (Sandrini et al., 2007; Roden et al., 2016; this study in 2004) and likely

### 322 5.2. Cant trends and variability

related to ventilation and water masses variability.

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Table 3.

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



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The long-term C<sub>T</sub> trend that we estimated in AABW in the eastern Enderby Basin (2.0±0.5 μmol.kg<sup>-1</sup>.decade<sup>-1</sup>) is slightly faster than the  $C_T$  trends estimated in the WSBW in the Weddell Gyre:  $+1.2\pm0.5~\mu mol.kg^{-1}.decade^{-1}$  over the period 1973-2011 and +1.6±1.4 µmol.kg<sup>-1</sup>.decade<sup>-1</sup> when restricted to 1996-2011 (van Heuven et al., 2014). Along the SR03 line (south of Tasmania) reoccupied in 1995, 2001, 2008 and 2011, Pardo et al (2017) evaluated a C<sub>T</sub> trend of +2.4±0.2 µmol.kg<sup>-1</sup>.decade<sup>-1</sup> in the AABW composed of ALBW and RSBW in this sector. This is higher than the C<sub>T</sub> trends found at our location and in the Weddell Gyre, but surprisingly, this was not associated with a significant increase in Cant. The CT trend in AABW along the SR03 section was likely due to the intrusion of old and C<sub>T</sub>-rich waters also revealed by an increase in silicate concentrations during 1995-2011 (Pardo et al., 2017). This is a clear example of decoupling between C<sub>T</sub> and C<sub>ant</sub> trends in deep and bottom waters as observed at our location in the last decade (Table 2). For C<sub>ant</sub>, our 40-years trend estimate (1.6±0.6 µmol.kg<sup>-1</sup>.decade<sup>-1</sup>) appears close to the trend reported by Rios et al. (2012) in the South-Western Atlantic AABW from 6 reoccupations between 1972 and 2003 (+1.5 µmol.kg<sup>-1</sup>.decade<sup>-1</sup>). However, if we limit our result to the period 1978-2002 or 1978-2004 (about the same period as in Rios et al., 2012), our trend is much larger (+3-4 µmol.kg<sup>-1</sup>.decade<sup>-1</sup>). At our location, the C<sub>ant</sub> trend over 40 years (+1.6±0.6 μmol.kg<sup>-1</sup>.decade<sup>-1</sup>) explains most of the observed C<sub>T</sub> increase (+2.0±0.5 µmol.kg<sup>-1</sup>.decade<sup>-1</sup>). The residual of +0.4 µmol.kg<sup>-1</sup>.decade<sup>-1</sup> reflects changes in natural processes affecting the carbon content (different AABW sources, ventilation, mixing with deep waters, remineralization or carbonates dissolution). Although this is a weak signal, the natural C<sub>T</sub> change (C<sub>Tnat</sub>) mirrors the observed decrease in O2 by -0.80±0.4 µmol.kg<sup>-1</sup>.decade<sup>-1</sup>. This O2 decrease detected in the Enderby Basin appears to be a real feature that was documented at large scale for 1960-2010 in deep SO basins (Schmidtko et al. 2017), suggesting that the changes observed at 56.5°S-63°E are related to large-scale processes, possibly due to a decrease in AABW formation (Purkey and Johnson, 2012).

## 5.3. Recent Cant stability

Although most studies suggest a gradual accumulation of Cant in the AABW, our time-series highlights significant pluriannual changes, in particular over the last decade when Cant concentrations were as low as around the year 2000 (Fig. 4a) and decoupled from the increase in C<sub>T</sub> (Fig. 4b). This result is difficult to interpret because at our location, away from AABW sources (Fig. 1), the temporal variability observed in the AABW layer can result from many remote processes occurring at the AABW formation sites (such as wind forcing, ventilation, sea-ice melting, thermodynamic, biological activity and air-sea exchanges), but internal processes during the transport of AABWs (such as organic matter remineralization, carbonate dissolution and mixing with surrounding waters) must also be taken into account. The apparent steady Cant feature suggests that AABWs found at our location has stored less Cant in recent years. This might be linked to reduced CO2 uptake in the AABW formation regions, as recognized at large-scale in the SO from the late 1980s to 2001 (Le Quéré et al., 2007; Metzl, 2009; Lenton et al., 2012; Landschützer et al., 2015). This large-scale response in the SO during a positive trend in the Southern Annular Mode (SAM) is mainly associated to stronger winds driven by accelerating greenhouse gas emissions and stratospheric ozone depletion, leading to warming and freshening in the SO (Swart et al., 2018), change in the ventilation of the carbon-rich deep waters and reduced CO2 uptake (Lenton et al., 2009). The reconstructed pCO2 fields by Landschützer et al. (2015) suggest that the reduced CO<sub>2</sub> sink in the 1990s is identified at high latitudes in the SO (see Fig. 2a and S9 in Landschützer et al., 2015). However, as opposed to the circumpolar open ocean 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 fCO2 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 fCO2 data are sparse, especially before 1990, and cruises were mainly 379 conducted in austral summer when the spatio-temporal fCO2 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 mixing. This leads to various estimates of the air-sea CO2 fluxes around Antarctica depending on the region and 381 382 period and large uncertainty when attempting to detect long-term trends (Gregor et al., 2018). In particular, in polynyas and AABW formation regions where fCO<sub>2</sub> is low and where katabatic winds prevail, 383 384 very strong instantaneous CO2 sink can occur at the local scale (up to -250 mmol C.m<sup>-2</sup>.d<sup>-1</sup> 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 fCO<sub>2</sub> in austral summer vary in a very large range (150-450 µatm), with the lowest fCO<sub>2</sub> observed in the shelf region generating very strong local CO<sub>2</sub> sink (-221 mmol C.m<sup>-2</sup>.d<sup>-1</sup> according to Roden et al. 387 2016). The carbon uptake was particularly enhanced near Cape Darnley and coincided with the highest Cant 388 389 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 fCO<sub>2</sub> values in 1993-1995 were as low as 100 µatm (Gibson and Trull, 1999) 391 leading to a strong local CO<sub>2</sub> uptake of -30 mmol C.m<sup>-2</sup>.d<sup>-1</sup> in summer. In addition, Roden et al. (2013) found a 392 large C<sub>T</sub> increase over 16 years (+34 μmol.kg<sup>-1</sup>) in the Prydz Bay, which is much higher than the anthropogenic 393 signal alone (+12 µmol.kg<sup>-1</sup>) 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<sub>T</sub> 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<sub>T</sub> and C<sub>ant</sub> 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 CO2 uptake in this region using the qualified fCO2 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 Cant 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 Cant 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 (Couldrey 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 Cant concentrations in AABW 412 observed at our location. As AABWs from different sources spread and mix with C<sub>T</sub>-rich deep waters before 413 reaching our location (Fig. 1), less AABW formation and export would result in an increase in C<sub>T</sub> (increase in 414 C<sub>Tnat</sub>) not associated with an increase in C<sub>ant</sub>, and a decrease in O<sub>2</sub> (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





 $416 \qquad contribution \ of \ older \ RSBW \ and/or \ ALBW \ that \ have \ lower \ C_{ant} \ and \ O_2 \ concentrations \ compared \ to \ CDBW \ formed$ 

The distribution and evolution of Cant in the bottom layer of the SO are related to complex interactions between

417 at Cape Darnley and Prydz Bay.

#### 6. Conclusion

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420 climatic forcing, air-sea CO<sub>2</sub> 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 Cant changes in AABW over 40 years in this region. 424 Our results suggest that the accumulation of Cant explains most of (but not all) the observed increase in C<sub>T</sub>. We 425 also detected a decrease in O<sub>2</sub> 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<sub>ant</sub> 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 Cant trends on short periods, with a maximum increase of 6.5 µmol.kg-1.decade-1 430 between 1987 and 2004 and an apparent stability in the last 20 years (despite an increase in C<sub>T</sub>). This suggests that 431 AABWs have stored less C<sub>ant</sub> 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<sub>2</sub> 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<sub>T</sub> or fCO<sub>2</sub> 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 Cant in AABW since 1998 could also be directly linked to a decrease in AABW formation in the 1990s (Purkey and Johnson, 2012) or a change in the contributions of 436 437 AABWs from different sources, especially in the Prydz Bay region (Williams et al., 2016). In these scenarios, an 438 increased contribution of C<sub>T</sub>-rich and O<sub>2</sub>-poor older CDW would also explain the decoupling between C<sub>ant</sub> and C<sub>T</sub> 439 (increase in C<sub>Tnat</sub>) and decrease in O<sub>2</sub> concentrations observed in recent years. The decoupling between C<sub>ant</sub> and 440 C<sub>T</sub> 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<sub>T</sub>-rich waters (Pardo et al., 2017). This highlights the importance of the ocean 442 circulation in influencing the temporal C<sub>T</sub> and C<sub>ant</sub> 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 Cant changes in deep and bottom ocean waters are still challenging, 445 as the C<sub>ant</sub> concentrations remain low compared to C<sub>T</sub> measurements accuracy (at best ± 2 μmol.kg<sup>-1</sup>, Bockmon 446 and Dickson, 2015) and uncertainties of data-based methods (± 6 µmol.kg<sup>-1</sup>). 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<sub>2</sub> 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 al., 2014; Ito et al., 2015, Yamamoto et al., 2015), it is important to maintain time-series observations to 452 453 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). OISO original data stations are available at:
- 462 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
- prepared the manuscript with contributions from CLM and NM.

# 467 Competing interests

The authors declare that they have no conflict of interest.

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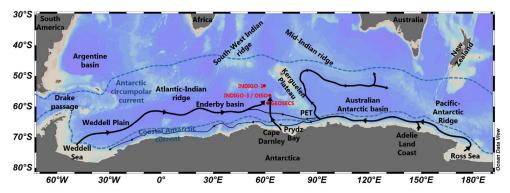




# 802 Table 1. List of the cruises used in this study.

Cruise	Station	Location	Year	Month
GEOSEC	CS 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	1 11	63.0°E / 56.5°S	1998	February
OISO-03	3 11	63.0°E / 56.5°S	1998	December
OISO-05	5 11	63.0°E / 56.5°S	2000	August
OISO-06	5 11	63.0°E / 56.5°S	2001	January
OISO-08	3 11	63.0°E/56.5°S	2002	January
OISO-11	1 11	63.0°E / 56.5°S	2004	January
OISO-18	3 11	63.0°E/56.5°S	2009	December
OISO-19	9 11	63.0°E / 56.5°S	2011	January
OISO-21	1 11	63.0°E / 56.5°S	2012	February
OISO-23	3 11	63.0°E / 56.5°S	2014	January
OISO-26	5 11	63.0°E / 56.5°S	2016	October
OISO-27	7 11	63.0°E / 56.5°S	2017	January
OISO-28	3 11	63.0°E/56.5°S	2018	January





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





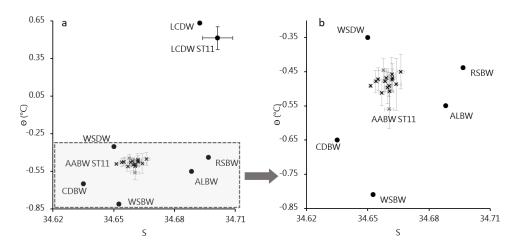


Figure 2. (a) Full Θ-S diagram of studied water masses and (b) zoomed on bottom waters. Values are from literature for the WSBW (Fukamachi et al., 2010; van Heuven, 2013; Pardo et al., 2014; Robertson et al., 2002), the WSDW (Carmack and Foster, 1975; Fahrbach et al., 1994; van Heuven, 2013; Robertson et al., 2002), the RSBW (Fukamachi et al., 2010; Gordon et al., 2015; Johnson, 2008; Pardo et al., 2014), the ALBW (Fukamachi et al., 2010; Johnson, 2008; Pardo et al., 2014), the CDBW (Ohshima et al., 2013) and the LCDW (Lo Monaco et al., 2005a; Pardo et al., 2014; Smith and Treguer, 1994), and from the OISO-ST11 dataset for the OISO-ST11 AABW and OISO-ST11 LCDW. Error bars are calculated from the individual annual averaged values for the OISO-ST11 AABW and from all data for the OISO-ST11 LCDW. For the OISO-ST11 AABW, the grey cross are the GEOSECS (lowest Θ) and INDIGO-1 (highest Θ)



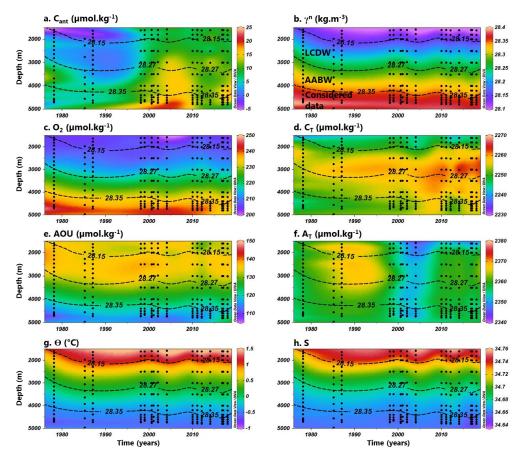


Figure 3. Hovmöller section of the carbon related properties parameters ( $C_{ant}$  via TrOCA,  $O_2$ , AOU,  $C_T$  and  $A_T$ ) and hydrological properties ( $\theta$ , S and  $\gamma^n$ ) of the dataset presented in Table 1 from 1978 to 2018 and from 1500 m to the bottom. Data points are represented by the black dots. The dash isolines represent the water masses separation by  $\gamma^n$  detailled on the  $\gamma^n$  plot. Figure produced with ODV (Schlitzer et al., 2019).

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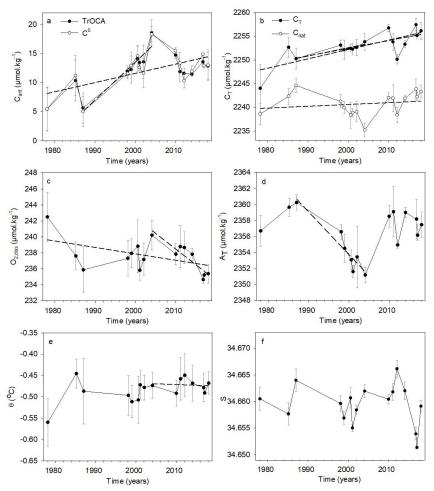


Figure 4. Interannual variability (full lines) and significant trends (at 95%, see Table 2; dotted lines) for the 40 years of observation of the OISO-ST11 AABW properties, including (a)  $C_{ant}$  by the TrOCA and the  $C^{\circ}$  method, (b)  $C_{T}$  and  $C_{nat}$ , (c)  $O_{2}$ , (d)  $A_{T}$ , (e)  $\Theta$  and (f) S.





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

Period	S	°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>	$A_T$ $\mu mol.kg^{-1}$	$C_T$ $\mu mol.kg^{-1}$	C <sub>ant</sub> TrOCA µmol.kg <sup>-1</sup>
1978-2018	-0.001 ± 0.001	$0.01 \pm 0.01$	-1.2 ± 0.9	$0.2 \pm 0.2$	-0.8 ± 0.4	0.7 ± 0.0	-0.1 ± 0.1	2.0 ± 0.5	1.6 ± 0.6
1987-2018	$-0.001 \pm 0.001$	$0.01 \pm 0.01$	$-1.9 \pm 1.4$	$0.3 \pm 0.4$	$-0.3 \pm 0.5$	$0.2 \ \pm \ 0.5$	$0.6 \ \pm 0.1$	$1.6 \ \pm 0.5$	$1.1 \ \pm 0.8$
1987-2004	$-0.003 \pm 0.002$	$0.01 \pm 0.01$	$\textbf{-6.5} \hspace{0.2cm} \pm \hspace{0.2cm} \textbf{1.8}$	$0.9 \pm 0.9$	$1.7\pm1.0$	$-1.7 \pm 1.0$	$\textbf{-5.3} \ \pm \textbf{0.1}$	$\textbf{1.8} \ \pm \textbf{0.4}$	$\textbf{6.5} \ \pm \textbf{1.0}$
2004-2018	$-0.006 \pm 0.003$	$\textbf{0.02} \ \pm \textbf{0.01}$	$-1.8 \pm 4.5$	$-0.5 \pm 1.0$	$-3.9 \pm 0.7$	$4.0 \ \pm 0.8$	$3.4 \pm 0.2$	$1.7 \pm 1.9$	$-3.5 \pm 1.5$

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

Source	Location	Water masses considered	Year	$C_{ant}$ $\mu mol.kg^{-1}$	
Pardo et al. (2014)	Averaged AABW	WSBW-RSBW-	1994	12	
Fig. 5	composition	ALBW	1,,,,,	12	
Lo Monaco et al. (2005b)	WOCE line I6	WSBW	1996	15	
Fig. 4b	(30°E; 50°-70°S)	CDBW		20	
Sandrini et al. (2007)	Ross Sea	HSSW Sea		Max. of 30	
Fig. 4a	Ross Sea	(previous RSBW)	2002/2003	Wax. of 50	
Shadwick et al. (2014)	Mertz polynya and	ALBW	2007/2008	15	
Table 2	Adelie depression	ALDW	2007/2008	13	
Roden et al. (2016)	South Indian ocean	WSDW-CDW-	2006	25	
Table 2	(30°-80°E; 60°-69°S)	CDBW			
van Heuven et al. (2011)	Weddell gyre	WCDW	2005	16	
Fig.13	(0°E; 55°-71°S)	WSBW	2005	10	
-			1978-1987	7 ± 3	
			1987-1998	9 ± 4	
This study	Enderby basin	WSDW-CDBW-	1987-2004	13 ± 4	
2 may	(56.5°S-63°E)	RSBW-ALBW	1998-2004	14 ± 2	
			2010-2018	$13 \pm 1$	
			1978-2018	$12 \pm 3$	

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