

# 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 the AABW, from the average concentration of 7 μmol.kg<sup>-1</sup> calculated for the period 1978-1987 to the average concentration of 13 μmol.kg<sup>-1</sup> for the period 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 total carbon (C<sub>T</sub>) and C<sub>ant</sub> increasing rates in the 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 C<sub>ant</sub> in the different formation regions. This is, however, modulated by significant interannual to multi-annual variability associated with variations in hydrological (potential temperature (Θ), salinity (S)) and biogeochemical (C<sub>T</sub>, total alkalinity (A<sub>T</sub>), dissolved oxygen (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 AABWs needs to be carefully considered on the extrapolated estimation of C<sub>ant</sub> sequestration based on sparse observations over several years.

## 1 Introduction

Carbon dioxide (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  $C_{\text{ant}}$  ocean sequestration (Matear, 2001; Orr et al., 2001; McNeil et al., 2003; Gruber et al., 2009; Khatiwala et al., 2009). Ocean  $C_{\text{ant}}$  uptake and sequestration have the benefit to limit the atmospheric  $\text{CO}_2$  increase but also result in a gradual decrease of the ocean pH (Gattuso and Hansson, 2011; Jiang et al., 2019). Understanding the oceanic  $C_{\text{ant}}$  sequestration and its variability is of major importance to predict future atmospheric  $\text{CO}_2$  concentrations, impact on the climate and impact of the pH change on marine ecosystems (de Baar, 1992; Orr et al., 2005; Ridgwell and Zeebe, 2005).

$C_{\text{ant}}$  in seawater cannot be measured directly and the evaluation of the relatively small  $C_{\text{ant}}$  signal from the total inorganic dissolved carbon ( $C_T$ ; around 3 %; Pardo et al., 2014) is still a challenge to overcome. Different approaches have been developed in the last 40 years to quantify  $C_{\text{ant}}$  concentrations in the oceans. The ‘historical’ back calculation method based on  $C_T$  measurement and preformed inorganic carbon estimate ( $C^0$ ) was independently published by Brewer (1978) and Chen and Millero (1979). This method has been often applied at regional and basin scale (Chen, 1982; Poisson and Chen, 1987; Chen, 1992; Goyet et al., 1998; Körtzinger et al., 1998, 1999; Lo Monaco et al., 2005a). More recently the TrOCA method (Tracer combining Oxygen, dissolved Carbon and total Alkalinity) has been developed (Touratier and Goyet, 2004a, b; Touratier et al., 2007) and applied in various regions including the SO (e.g. Lo Monaco et al., 2005b; Sandrini et al., 2007; Van Heuven et al., 2011; Pardo et al., 2014; Shadwick et al., 2014; Roden et al., 2016; Kerr et al., 2018). Comparisons with other data-based methods show significant differences in  $C_{\text{ant}}$  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 is a need to better explore the  $C_T$  and  $C_{\text{ant}}$  temporal variability in the deep ocean, especially in the SO where observations are relatively sparse.

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

The ability of AABW to accumulate  $C_{\text{ant}}$  has been controversial since one can believe that the ice coverage limits the invasion of  $C_{\text{ant}}$  in Antarctic surface waters (e.g. Poisson and Chen, 1987). This is, however, not the case in polynyas, and several studies have reported significant  $C_{\text{ant}}$  signals in AABW formation regions, likely due to the uptake of  $\text{CO}_2$  induced by high primary production (Sandrini et al., 2007; van Heuven et al., 2011, 2014; Shadwick et al., 2014; Roden et al., 2016). However, little is known about the variability and evolution of the  $\text{CO}_2$  fluxes in

AABW formation regions, and since biological and physical processes are strongly impacted by seasonal and interannual climatic variations (Fukamachi et al., 2000; Gordon et al., 2010, McKee et al., 2011; Gordon et al., 2015; Gruber et al., 2019b), the amount of  $C_{ant}$  stored in the AABWs may be very variable, which could bias the estimates of  $C_{ant}$  trends derived from data sets collected several years apart (e.g. Williams et al., 2015; Pardo et al., 2017; Murata et al., 2019).

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

## 2 Studied area

### 2.1 AABW sampling during the last 40 years

Most of the data used in this study were obtained in the frame of the long-term observational project OISO (Ocean Indien Service d'Observations) conducted since 1998 onboard the R.S.V. Marion-Dufresne (IPEV/TAAF). During these cruises, several stations are visited, but only one station is sampled down to the bottom (4800 m) south of the Polar Front at 63.0° E and 56.5° S (hereafter noted OISO-ST11). This station is located in the Enderby Basin on the Western side of the Kerguelen Plateau (Fig. 1) and coincides with the station 75 of the INDIGO-3 cruise (1987). In our analysis, we also included data from the station 14 (deepest sample taken at 5109 m) of the INDIGO-1 cruise (1985) and the station 430 (deepest sample taken at 4710 m) of the GEOSECS cruise (1978) located near OISO-ST11 sampling site (405 km and 465 km away from it, respectively; Fig. 1). All the re-occupations used in this analysis are listed in Table 1. Since seasonal variations are only observed in the surface mixed layer (Metzl et al., 2006), we used the observations available for all seasons (Table 1).

Table 1

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

The circulation in the SO is mainly governed by the Antarctic Circumpolar Current (ACC) that flows eastward, while the Coastal Antarctic Current (CAC) flows westward (Fig. 1). The ACC and the CAC influence the circulation of the entire water column, including the AABWs. The main AABW formation sites are the Weddell Sea, where Weddell Sea Deep Water and Weddell Sea Bottom Water are produced (WSDW and WSBW, respectively; Gordon, 2001; Gordon et al., 2010), the Ross Sea for the Ross Sea Bottom Water (RSBW; Gordon et al., 2009, 2015), the Adelie Land coast for the Adelie Land Bottom Water (ALBW; Williams et al., 2008, 2010) and the Cape Darnley Polynya for the Cape Darnley Bottom Water (CDBW; Ohshima et al., 2013). AABW formation has also been observed in the Prydz Bay (Yabuki et al., 2006; Rodehacke et al., 2007). There, three polynyas and two ice shelves have been identified as Prydz Bay Bottom Water (PBBW) production hotspots from seal tagging data (Williams et al., 2016). This PBBW flows out the Prydz Bay through the Prydz Channel and get mixed with the CDBW. The mix of CDBW and PBBW (hereafter called CDBW) represents a significant AABW export (13 % of all AABWs exports; Ohshima et al., 2013).

The largest bottom water source of the global ocean is the Weddell Sea (Gordon et al., 2001). The exported WSDW is a mixture of the WSBW and Warm Deep Water (WDW). The WDW is a slightly modified Low Circumpolar Deep Water (LCDW) by mixing with High Salinity Surface Water (HSSW) when the LCDW enters the Weddell

basin (see Fig. 2 in van Heuven et al., 2011). The WSDW mixes with the LCDW during its transit from the Weddell basin. A part of the WSDW deflecting southward with the ACC in the Enderby Basin reaches the north-western part of the Princess Elizabeth Trough (PET) region (area separating the Kerguelen Plateau from the Antarctic continent), where it mixes with other types of AABWs (Heywood et al., 1999; Orsi et al., 1999). The PET deepest point is 3750 m, deep enough to allow AABWs to flow between the Australian Antarctic Basin and the Enderby Basin (Heywood et al., 1999).

At the east of the PET, the CAC transports a mixture of RSBW and ALBW and accelerates northward along the eastern 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 by the southern part of the PET (Heywood et al., 1999; Orsi et al., 1999; Van Wijk and Rintoul, 2014) and mixes with the CDBW. The mixture of CDBW and ALBW-RSBW either flows westward with the CAC and dilutes with the LCDW (Meijers et al., 2010) or flows northward (Ohshima et al., 2013) and mixes with the WSDW before reaching the location of our time-series station in the eastern Enderby Basin.

Figure 1

### 2.3 AABW definition

The distinction of water masses is usually performed according to neutral density ( $\gamma^n$ ) layers. In the SO, LCDW and AABW properties are generally well defined in the range 28.15-28.27 kg.m<sup>-3</sup> and 28.27-bottom, respectively (Orsi et al., 1999; Murata et al 2019). However, to interpret the long-term variability of the properties in the AABW core at our location, we prefer to adjust the AABW definition to a narrow (more homogeneous) layer that we call Lower Antarctic Bottom Water (LAABW), characterised by  $\gamma^n > 28.35$  kg.m<sup>-3</sup> (roughly ranging from 4200m to 4800m, see Fig. 3). This definition corresponds to the AABW characteristics observed at higher latitudes in the Indian SO sector (Roden et al., 2016). The layer above the LAABW is hereafter called Upper Antarctic Bottom Water (UAABW).

## 3 Material and methods

### 3.1 Validation of the data

For 1998-2004, the OISO data were quality controlled in CARINA (Lo Monaco et al., 2010) and for 2005 and 2009-2011 in GLODAPv2 (Key et al., 2015; Olsen et al., 2016, 2019). The 3 additional datasets from GEOSECS, INDIGO-1 and INDIGO-3 were first qualified in GLODAPv1 (Key et al., 2004) and used for the first  $C_{ant}$  estimates in the Indian Ocean (Sabine et al., 1999). The adjustments recommended for these historical datasets have been revisited in CARINA and GLODAPv2. In this paper we used the revised adjustments applied to the GLODAPv2 data product, with one exception for the total alkalinity ( $A_T$ ) data from INDIGO-3 for which we applied an intermediate adjustment between the recommendation from GLODAPv1 (confirmed in CARINA) for no adjustment and the adjustment by -8  $\mu\text{mol.kg}^{-1}$  applied to the GLODAPv2 data product (justification in Supp. Mat.).

For the recent OISO cruises conducted in 2012-2018 not yet included in the most recent GLODAPv2 product, we have proceeded to a data quality control in deep waters where  $C_{ant}$  concentrations are low and subject to very small changes from year to year (see Supp. Mat.).

### 3.2 Biogeochemical measurements

Measurement methods during OISO cruises were previously described (Jabaud-Jan et al., 2004; Metzl et al., 2006). In short, measurements were obtained using Conductivity-Temperature-Depth (CTD) casts fixed on a 24 bottles rosette equipped with 12 L General Oceanics Niskin bottles. Potential temperature ( $\Theta$ ) and salinity (S) measurements have an accuracy of 0.002 °C and 0.005 respectively.  $A_T$  and  $C_T$  were sampled in 500 mL glass bottles and poisoned with 100  $\mu$ L of mercuric chloride saturated solution to halt biological activity. Discrete  $C_T$  and  $A_T$  samples were analyzed onboard by potentiometric titration derived from the method developed by Edmond (1970) using a closed cell. The repeatability 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 in bottom waters). The accuracy of  $C_T$  and  $A_T$  measurements was ensured by daily analyses of Certified Reference Materials (CRMs) provided by A.G. Dickson laboratory (Scripps Institute of Oceanography). Dissolved oxygen ( $O_2$ ) concentration 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). The thiosulphate solution used for the Winkler titration was calibrated using iodate standard solution (provided by Ocean Scientific International Limited) to ensure the standard  $O_2$  accuracy of 2  $\mu$ mol.kg<sup>-1</sup>. Nitrate ( $NO_3$ ) and Silicate (Si) concentrations were measured onboard or onshore 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.  $NO_3$  data are not available for all the cruises used in this analysis. The mean  $NO_3$  concentrations in the LAABW at OISO-ST11 is  $32.8 \pm 1.2$   $\mu$ mol.kg<sup>-1</sup> while the average value derived from the GLODAP-v2 database in bottom waters south of 50°S in the South Indian Ocean is  $32.4 \pm 0.6$   $\mu$ mol.kg<sup>-1</sup>. The lack of  $NO_3$  data for few cruises has been palliated by considering a climatological value of 33  $\mu$ mol.kg<sup>-1</sup> with a limited impact on  $C_{ant}$  determined by the  $C^\circ$  method ( $<2$   $\mu$ mol.kg<sup>-1</sup> on estimates based on the differences observed between  $NO_3$  measurements and the climatological value).

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

The TrOCA method was first presented by Touratier and Goyet (2004a, b) and revised by Touratier et al. (2007). Following the concept of the quasi-conservative tracer NO (Broecker, 1974), TrOCA is a tracer defined as a combination of  $O_2$ ,  $C_T$  and  $A_T$ , following:

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

where  $a$  is defined in Touratier et al. (2007) as combination of the Redfield equation coefficients for  $CO_2$ ,  $O_2$ ,  $HPO_4^{2-}$  and  $H^+$ . For more details about the definition and the calibration of this parameter, please refer to Touratier et al. (2007). The temporal change in TrOCA is independent of biological processes and can be attributed to anthropogenic carbon (Touratier and Goyet, 2004a). Therefore,  $C_{ant}$  can be directly calculated from the difference between TrOCA and its pre-industrial value  $TrOCA^0$ :

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

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

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

In these expressions, coefficients a, b, c and d were adjusted by Touratier et al. (2007) from free anthropogenic CO<sub>2</sub> deep waters using the tracers Δ<sup>14</sup>C and CFC-11 from the GLODAPv1 database (Key et al., 2004). The final expression used to calculate C<sub>ant</sub> is:

$$C_{ant} = \frac{0.2 + 1.279 \left( C_T - \frac{1}{2} A_T \right) - e^{\left[ \frac{7.511 - (1.087 \cdot 10^{-2}) \cdot \theta - \frac{7.81 \cdot 10^5}{A_T^2} \right]}}{1.279}, \quad (4)$$

The consideration of the errors on the different parameters involved in the TrOCA method results in an uncertainty of ±6.25 μmol.kg<sup>-1</sup> (mostly due to the parameter a, leading to ±3.31 μmol.kg<sup>-1</sup>). As this error is relatively large compared to the expected C<sub>ant</sub> concentrations in deep and bottom SO waters (Pardo et al., 2014) we will compare the TrOCA results using another indirect method to interpret C<sub>ant</sub> changes over 40 years.

### 3.4 C<sub>ant</sub> calculation using the preformed inorganic carbon (C<sup>0</sup>) method

To support the C<sub>ant</sub> trend determined with the TrOCA method, C<sub>ant</sub> was also estimated using a back-calculation approach noted C<sup>0</sup> (Brewer, 1978; Chen and Millero, 1979), previously adapted for C<sub>ant</sub> estimates along the WOCE-I6 section between South Africa and Antarctica (Lo Monaco et al., 2005a). This method consists in the correction of the measured C<sub>T</sub> for the biological contribution (C<sub>bio</sub>) and the preindustrial preformed C<sub>T</sub> (C<sup>0</sup><sub>PI</sub>):

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

C<sub>bio</sub> (Eq. 6) depends on carbonate dissolution and organic matter remineralization, taking account of the corrected C/O<sub>2</sub> ratio from Kortzinger et al. (2001):

$$C_{bio} = 0.54 \Delta A_T - (C/O_2 + 0.5N/O_2) \Delta O_2, \quad (6)$$

Where C/O<sub>2</sub> = 106/138 and N/O<sub>2</sub> = 16/138. ΔA<sub>T</sub> and ΔO<sub>2</sub> are the difference between the measured values (A<sub>T</sub> and O<sub>2</sub>) and the preformed values (A<sub>T</sub><sup>0</sup> and O<sub>2</sub><sup>0</sup>). A<sub>T</sub><sup>0</sup> (Eq. 7) has been computed by Lo Monaco et al. (2005a) as a function of Θ, S and the conservative tracer PO:

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

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 as follows (the N/P ratio of 16 is from Anderson and Sarmiento, 1994):

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

To determine O<sub>2</sub><sup>0</sup>, it is assumed that the surface water is in full equilibrium with the atmosphere (O<sub>2</sub><sup>0</sup>=O<sub>2,sat</sub>; Benson and Krause, 1980) and that after subduction O<sub>2</sub> in a given water mass is only impacted by the biological activity (Weiss, 1970). A correction of O<sub>2</sub><sup>0</sup> has been proposed by Lo Monaco et al. (2005a) to take account of the undersaturation of O<sub>2</sub> due to sea-ice cover at high latitudes. O<sub>2</sub><sup>0</sup> 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<sub>2</sub> undersaturation in ice-covered surface waters α = 12 % (Anderson et al., 1991) according to Eq. 9:

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

C<sup>0</sup><sub>PI</sub> in equation 5 is a function of the current preformed C<sub>T</sub> (C<sup>0</sup><sub>obs</sub>) and a reference water term (Eq. 10):

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

C<sub>0,obs</sub> has been computed similarly as A<sub>T</sub><sup>0</sup> (Eq. 11):

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

Where the reference water term is a constant for a given time of observation, corresponding to the time when  $C^0_{\text{obs}}$  is parameterized. In this paper, we used the parameterization given by Lo Monaco et al., (2005a) and their estimated value for the reference term of  $51 \mu\text{mol.kg}^{-1}$ . This number has been computed using an optimum multiparametric (OMP) model to estimate the mixing ratio of the North Atlantic deep water in the SO (used as reference water, i.e. old water mass where  $C_{\text{ant}} = 0$ ). For more details about the  $C^0$  method, which has a final error of  $\pm 6 \mu\text{mol.kg}^{-1}$ , please see Lo Monaco et al. (2005a).

## 4 Results

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 2. The LCDW layer ( $\gamma^n = 28.15\text{-}28.27 \text{ kg.m}^{-3}$ ) is characterized by minimum  $\text{O}_2$  concentrations (Fig. 2c), higher  $C_T$  (Fig. 2b) and lower  $C_{\text{ant}}$  concentrations than in the AABW (Fig. 2a).  $C_{\text{ant}}$  concentrations were not significant in the LCDW until the end of the 1990s ( $<6 \mu\text{mol.kg}^{-1}$ ), then our data show an increase in  $C_{\text{ant}}$  between the two 1998 reoccupations, followed by relatively constant  $C_{\text{ant}}$  concentrations ( $10 \pm 3 \mu\text{mol.kg}^{-1}$ ). In the LAABW ( $\gamma^n > 28.35 \text{ kg.m}^{-3}$ ), well identified by low  $\Theta$ , low  $S$  and high  $\text{O}_2$ ,  $C_{\text{ant}}$  concentrations are higher than in the overlying UAABW and LCDW (Fig. 2a). The evolutions of the mean properties in the LAABW over 40 years are shown in Fig. 3. In this layer,  $C_{\text{ant}}$  concentrations increased from  $5 \pm 4 \mu\text{mol.kg}^{-1}$  in 1978 and  $7 \pm 4 \mu\text{mol.kg}^{-1}$  in the mid-1980s to  $13 \pm 2 \mu\text{mol.kg}^{-1}$  at the end of the 1990s and up to  $19 \pm 2 \mu\text{mol.kg}^{-1}$  in 2004 (Fig. 3a). Figure 3a also shows a very good agreement between the TrOCA method and the  $C^0$  method for both the magnitude and variability of  $C_{\text{ant}}$  in the LAABW. Our results show a mean  $C_{\text{ant}}$  trend in the LAABW of  $+1.4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  over the full period and a maximum trend of the order of  $+5.2 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  over 1987-2004 (Table 2). These trends are lower than the theoretical trend expected from the increase in atmospheric  $\text{CO}_2$ . Indeed, assuming that the surface ocean  $f\text{CO}_2$  follows the atmospheric growth rate ( $+1.8 \mu\text{atm.year}^{-1}$  over 1978-2018), the theoretical  $C_{\text{ant}}$  trend at the AABW formation sites would be of the order of  $+8 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ . The observed slow  $C_{\text{ant}}$  trends can be partly explained by the transit time for AABW to reach our study site and the mixing of AABWs with older LCDW that contain less  $C_{\text{ant}}$  over their transit (Fig. 2a).

### Figure 2

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 accumulation of  $C_{\text{ant}}$  (Table 2). Our data also show a significant decrease in  $\text{O}_2$  concentrations by  $0.8 \pm 0.4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  over the 40-years period (Fig. 3c, Table 2) that could be caused by reduced ventilation, as suggested by Schmidtko et al. (2017) who observed significant  $\text{O}_2$  loss in the global ocean. In the deep Indian SO sector, these authors found a trend approaching  $-1 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  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  $A_T$ . The low  $A_T$  values observed over 2000-2004 (Fig. 3d) could suggest reduced calcification in the upper ocean leading to less sinking of calcium carbonate tests and a decrease in  $A_T$  in deep and bottom waters over this period (Fig. 2d). For this period the increase in  $C_T$  was lower than the accumulation of  $C_{\text{ant}}$ , but such feature is disputable in view of the uncertainty on the  $C_{\text{ant}}$  calculation. This event is followed by an increase in the ‘natural’ component of  $C_T$  ( $C_{\text{nat}}$ , calculated as the difference between  $C_T$  and  $C_{\text{ant}}$ ) since 2004 associated to a decrease in  $\text{O}_2$  and no increase in  $C_{\text{ant}}$  (Table 2). These trends were not associated with a significant trend in  $\theta$  or  $S$  (Fig. 3e,f, Table 2). The increase in  $C_{\text{nat}}$  is thus unlikely originating from increased mixing with LCDW during bottom waters

transport, confirming that our LAABW definition exclude mixing with the LCDW. Enhanced organic matter remineralization is also unlikely since  $\text{NO}_3$  did not show any significant trend (Table 2).

Table 2

Figure 3

Importantly, our data show substantial interannual variations in LAABW properties, which could significantly impact the trends estimated from limited reoccupations (e.g. Williams et al., 2015; Pardo et al., 2017; Murata et al., 2019). For example, we found relatively higher  $C_{\text{ant}}$  concentrations in 1985 ( $10 \mu\text{mol.kg}^{-1}$ ) compared to 1978 ( $5 \mu\text{mol.kg}^{-1}$ ) and 1987 ( $7 \mu\text{mol.kg}^{-1}$ ). This is linked to a signal of low  $S$  in 1985 (Fig. 3f) that could be due to a larger contribution of fresher waters such as the WSDW or CDBW. This could also be related to the different sampling locations. Over the last decade (2009-2018), our data show large and rapid changes in  $S$  that are partly reflected on  $C_T$  and  $\text{O}_2$ , and that could explain the relatively low  $C_{\text{ant}}$  concentrations observed over this period. Indeed, the  $S$  maximum observed in 2012 (correlated to higher  $\theta$ ) is associated with a marked  $C_T$  minimum (surprisingly almost as low as in 1987), as well as low  $A_T$  (hence low  $C_{T\text{nat}}$ ), and low  $\text{NO}_3$  concentrations. Since these anomalies were associated with a decrease in  $C_{\text{ant}}$  concentrations, one may argue for an increased contribution of bottom waters ventilated far away from our study site. A few years later our data show a  $S$  minimum (correlated to lower  $\theta$ ), associated with a rapid increase in  $C_T$  and a rapid decrease in  $\text{O}_2$  between 2013 and 2016, suggesting the contribution of a closer AABW type such as the CDBW. The freshening of  $-0.006 \text{ decade}^{-1}$  in  $S$  between 2004 and 2018 that we observed on the western side of the Kerguelen Plateau was also observed on the eastern side of the Plateau by Menezes et al. (2017) over a similar period. In this region, Menezes et al. (2017) evaluated a change in  $S$  by about  $-0.008 \text{ decade}^{-1}$  from 2007 to 2016 (against  $-0.002 \text{ decade}^{-1}$  between 1994 and 2007), suggesting an acceleration of the AABW freshening in recent years. However, they also reported a warming by  $+0.06 \text{ }^\circ\text{C.decade}^{-1}$ , while we observed cooler temperature in 2016-2018. This suggests that we sampled a different mixture of AABWs.

Figure 4

## 5 Discussion

### 5.1 LAABW composition at OISO-ST11

At each formation site, AABWs experienced significant temporal property changes, mostly recognized at decadal scale (e.g. freshening in the South Indian Ocean, Menezes et al., 2017) with potential impact on carbon uptake and  $C_{\text{ant}}$  concentrations during AABW formation (Shadwick et al., 2013). The  $\Theta$ - $S$  diagram constructed from yearly averaged data in bottom waters (Fig. 4) shows that the LAABW at OISO-ST11 is a complex mixture of WSDW, CDBW, RSBW and ALBW. The coldest type of LAABW was observed at the GEOSECS station at  $60^\circ \text{ S}$  ( $-0.56 \text{ }^\circ\text{C}$ ), while the warmer type of LAABW observed at the INDIGO-1 station at  $53^\circ \text{ S}$  ( $-0.44 \text{ }^\circ\text{C}$ ). These extreme  $\Theta$  values could be a natural feature or may be related to specific sampling. For the other cruises,  $\Theta$  in LAABW ranges from  $-0.51$  to  $-0.45 \text{ }^\circ\text{C}$  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. 1 and 4) and the significant  $C_{\text{ant}}$  concentrations that we calculated in the LAABW (Fig. 3a), the main contribution at our location is likely the younger and colder CDBW for which relatively high  $C_{\text{ant}}$  concentrations



have been recently documented (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 LCDW along the Antarctic shelf and the continental slope between 80° E and 30° E (Meijers et al., 2010; Roden et al., 2016). On the western side of the Kerguelen Plateau, CDBW also mixes with RSBW and ALBW (Orsi et al., 1999; Van Wijk and Rintoul, 2014). In this context, the  $C_{ant}$  concentrations observed in the bottom layer at OISO-ST11 are probably not linked to one single AABW source, but are likely a complex interplay of AABWs from different sources with different biogeochemical properties.

## 5.2 $C_{ant}$ concentrations

In order to compare our  $C_{ant}$  estimates with other studies, we separated the 40-years time-series into 3 periods: the first period (1978-1987) corresponds to historical data when  $C_{ant}$  is expected to be low; the second period (1998-2004) starts when the first OISO cruise was conducted (and using CRMs for  $A_T$  and  $C_T$  measurements) and lasts when  $C_{ant}$  concentrations in the LAABW are maximum (Fig. 3a); the third period consists in the observations performed in late 2009 to 2018 when the observed variations are relatively large for  $S$  and small for  $C_{ant}$ . The mean  $C_{ant}$  concentrations for each period are 7, 14 and 13  $\mu\text{mol.kg}^{-1}$ , respectively, which is consistent with the results from other studies (Table 3). The  $C_{ant}$  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 GLODAP and CARINA (Key et al., 2004; Lo Monaco et al., 2010; Olsen et al., 2016). In addition, the different methods used to estimate  $C_{ant}$  can lead to different results, especially in deep and bottom waters of the SO (Vázquez-Rodríguez et al., 2009). Overall, Table 3 confirms that  $C_{ant}$  concentrations were low in the 1970s 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 data-based estimates (Gruber, 1998; Sabine et al., 2004; Waugh et al., 2006; Khatiwala et al., 2013).

The observations presented in this analysis, although regional, offer a complement to recent estimates of  $C_{ant}$  changes evaluated between 1994 and 2007 in the top 3000 m for the global ocean (Gruber et al., 2019a). In the Enderby Basin at the horizon 2000-3000 m, the accumulation of  $C_{ant}$  from 1994 to 2007 is not uniform and ranges between 0 and 8  $\mu\text{mol.kg}^{-1}$  (Gruber et al., 2019a). At our station, in the LCDW (2000-3000m) the  $C_{ant}$  concentrations were not significant in 1978-1987 ( $-2$  to  $5 \mu\text{mol.kg}^{-1}$ ) but increase to an average of  $9 \pm 3 \mu\text{mol.kg}^{-1}$  in 1998-2018 (Fig. 2a), probably due to mixing with AABWs that contain more  $C_{ant}$ . Interestingly, this value is close but in the high range of the  $C_{ant}$  accumulation estimated from 1994 to 2007 in deep waters of the south Indian Ocean (Gruber et al., 2019a).

Not surprisingly, high  $C_{ant}$  concentrations are detected in the AABW formation regions (Table 3). The highest  $C_{ant}$  concentrations in bottom waters (up to 30  $\mu\text{mol.kg}^{-1}$ ) were observed in the ventilated shelf waters in the Ross Sea (Sandrini et al., 2007). In the Adélie and Mertz Polynya regions, Shadwick et al. (2014) observed high  $C_{ant}$  concentrations in the subsurface shelf waters (40-44  $\mu\text{mol.kg}^{-1}$ ) but lower values in the ALBW (15  $\mu\text{mol.kg}^{-1}$ ) due to mixing with older LCDW. In WSBW, all  $C_{ant}$  concentrations estimated from observations between 1996 and 2005 and with the TrOCA method (Table 3) lead to about the same values ranging between 13 and 16  $\mu\text{mol.kg}^{-1}$  (Lo Monaco et al., 2005b; van Heuven et al., 2011). In bottom waters formed near the Cape Darnley (CDBW), Roden et al. (2016) estimated high  $C_{ant}$  concentrations in bottom waters (25  $\mu\text{mol.kg}^{-1}$ ) resulting from the shelf

waters that contain very high amounts of  $C_{\text{ant}}$  ( $50 \mu\text{mol.kg}^{-1}$ ). The comparison with other studies confirms that far from the AABW formation sites, contemporary  $C_{\text{ant}}$  concentrations are not exceeding  $16 \mu\text{mol.kg}^{-1}$  on average. Table 3.

### 5.3 $C_{\text{ant}}$ trends and variability

Comparison of long-term  $C_{\text{ant}}$  trends in deep and bottom waters of the SO is limited to very few regions where repeated observations are available. To our knowledge, only 3 other studies evaluated the long-term  $C_{\text{ant}}$  trends in the SO 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  $C_T$  and  $C_{\text{ant}}$  have also been investigated in other SO regions, but limited to 2 to 4 reoccupations (Williams et al., 2015; Pardo et al., 2017; Murata et al., 2019). Given the  $C_{\text{ant}}$  variability depicted at our location (Fig. 3a), different trends can be deduced from limited reoccupations. As an example, Murata et al., (2019) evaluated the change in  $C_{\text{ant}}$  from data collected 17 years apart (1994–1996 and 2012–2013) along a transect around  $62^\circ \text{S}$  and found a 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 observation given that we found a minimum in  $C_{\text{ant}}$  concentrations between 2011 and 2014 (Fig. 3a) associated with a marked  $C_T$  minimum (Fig. 3b). In addition, our results show that the detection of  $C_{\text{ant}}$  trends appears very sensitive to the time period considered (Table 2). As an extreme case, the  $C_{\text{ant}}$  trend calculated for the period 1987–2004 is  $+5.2 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  (relatively close to the theoretical  $C_{\text{ant}}$  trend of  $+8 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ), but it reverses to  $-3.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  for the period 2004–2018.

The long-term  $C_T$  trend that we estimated in the LAABW in the eastern Enderby Basin ( $2.0 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ) is 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 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). Along the SR03 line (south of Tasmania) reoccupied in 1995, 2001, 2008 and 2011, Pardo et al. (2017) calculated 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 higher than the  $C_T$  trends found at our location and in the Weddell Gyre, but surprisingly, this was not associated with a significant increase in  $C_{\text{ant}}$ . The  $C_T$  trend in AABW along the SR03 section was likely due to the intrusion of old and  $C_T$ -rich waters also revealed by an increase in Si concentrations during 1995–2011 (Pardo et al., 2017). This is a clear example of decoupling between  $C_T$  and  $C_{\text{ant}}$  trends in deep and bottom waters as observed at our location in the last decade (Table 2). For  $C_{\text{ant}}$ , our 40-years trend estimate ( $1.4 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ) appears close to the trend reported by Rios et al. (2012) in the south-western Atlantic AABW from 6 reoccupations between 1972 and 2003 ( $+1.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ). 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 ( $+3\text{--}4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ).

At our location, the  $C_{\text{ant}}$  trend over 40 years ( $+1.4 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ) explains most of the observed  $C_T$  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 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  $C_T$  change ( $C_{\text{nat}}$ ) mirrors the observed decrease in  $\text{O}_2$  by  $-0.8 \pm 0.4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ . This  $\text{O}_2$  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 (Schmidt et al., 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 decrease in AABW formation (Purkey and Johnson, 2012).

#### 5.4 Recent $C_{\text{ant}}$ stability

Although most studies suggest a gradual accumulation of  $C_{\text{ant}}$  in the AABW, our time-series highlights significant multi-annual changes, in particular over the last decade when  $C_{\text{ant}}$  concentrations were as low as around the year 2000 (Fig. 3a) and decoupled from the increase in  $C_T$  (Fig. 3b). This result is difficult to interpret because at our location, away from AABW sources (Fig. 1), the temporal variability observed in the LAABW 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). Additionally, 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  $C_{\text{ant}}$  feature suggests that AABWs found at our location has stored less  $C_{\text{ant}}$  in recent years. This might be linked to reduced  $\text{CO}_2$  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  $C_T$ -rich deep waters and reduced  $\text{CO}_2$  uptake (Lenton et al., 2009). The reconstructed  $\text{pCO}_2$  fields by Landschützer et al. (2015) suggest that the reduced  $\text{CO}_2$  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 surface  $\text{fCO}_2$  and carbon uptake deduced from direct observations are not clearly identified in the seasonal ice zone (SIZ) and shelves around Antarctica, and thus in the AABW formation regions of interest to interpret our results (Laruelle et al., 2018). There, surface  $\text{fCO}_2$  data are sparse, especially before 1990, and cruises were mainly conducted in austral summer when the spatio-temporal  $\text{fCO}_2$  variability is very large and driven by multiple processes at regional or small scales, such as primary production, sea-ice formation and retreat, and water circulation and mixing. This leads to various estimates of the air-sea  $\text{CO}_2$  fluxes around Antarctica depending on the region and period and large uncertainty when attempting to detect long-term trends (Gregor et al., 2018).

In particular, in polynyas and AABW formation regions where  $\text{fCO}_2$  is low and where katabatic winds prevail, 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 the Ross Sea according to De Jong and Dunbar, 2017). In the Prydz Bay region where CDBW is formed, recent studies show that surface  $\text{fCO}_2$  in austral summer vary over a very large range ( $150\text{--}450 \text{ }\mu\text{atm}$ ), with the lowest  $\text{fCO}_2$  observed in the shelf region generating very strong local  $\text{CO}_2$  sink ( $-221 \text{ mmol C.m}^{-2}.\text{d}^{-1}$ ; Roden et al. 2016). The carbon uptake was particularly enhanced near Cape Darnley and coincided with the highest  $C_{\text{ant}}$  concentrations that Roden et al. (2016) estimated in the dense shelf waters that subduct to form AABW. In the Prydz Bay coastal region, surface  $\text{fCO}_2$  values in 1993-1995 were as low as  $100 \text{ }\mu\text{atm}$  (Gibson and Trull, 1999) 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 large  $C_T$  increase over 16 years ( $+34 \text{ }\mu\text{mol.kg}^{-1}$ ) in the Prydz Bay, which is much higher than the anthropogenic signal alone ( $+12 \text{ }\mu\text{mol.kg}^{-1}$ ) and likely explained by changes in primary production that would have been stronger in 1994. To our knowledge, this is the only direct observation of decadal  $C_T$  change in surface waters in a region of AABW formation (here the Prydz Bay) and it highlights the difficulty not only to evaluate the  $C_T$  and  $C_{\text{ant}}$  long-term trends in these regions but also to separate natural and anthropogenic signals when this water reaches the deep ocean. We attempted to detect long-term changes in  $\text{CO}_2$  uptake in this region using the qualified  $\text{fCO}_2$  data available in the

SOCAT database (Bakker et al., 2016), but our estimates (not shown) were highly uncertain due to very large spatial and temporal variability. To conclude, all previous studies conducted near or in AABW formation sites clearly reveal that these regions are potentially strong carbon sinks, but how the sink changed over 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 in the LAABW at our location is directly linked to the weakening of the carbon sink that was recognized at large-scale in the SO from the 1980s to mid-2000s (Le Quéré et al., 2007; Landschützer et al., 2015).

Changes in the accumulation of  $C_{\text{ant}}$  in AABW could also be directly related to changes in physical processes occurring in AABW formation regions. Decadal decreasing of sea-ice production and melting of sea-ice have been documented in several regions including Cape Darnley polynyas (Tamura et al., 2016; Williams et al., 2016). The consequent changes in Antarctic surface waters properties are transmitted into the deep ocean, notably the well-recognized freshening of AABWs over the last decades (Rintoul, 2007). The warming of bottom waters was also documented in the Enderby basin (Couldrey et al., 2013) as well as at a larger scale in all deep SO basins (Purkey and Johnson, 2010; Desbruyères et al., 2016). Associated to a decrease in AABW formation in the 1990s (Purkey and Johnson, 2012), these physical changes could explain the recent stability of  $C_{\text{ant}}$  concentrations in AABW observed at our location. As AABWs from different sources spread and mix with  $C_T$ -rich deep waters before reaching our location (Fig. 1), less AABW formation and export would result in an increase in  $C_T$  (increase in  $C_{\text{nat}}$ ) not associated with an increase in  $C_{\text{ant}}$ , and a decrease in  $O_2$  (as observed in recent years in Fig. 3a,b,c). Finally, it is also possible that the LAABW observed in recent years at our location is the result of a larger contribution of older RSBW, ALBW or even WSBW that have lower  $C_{\text{ant}}$  and  $O_2$  concentrations compared to CDBW formed at Cape Darnley and Prydz Bay.

## 6 Conclusion

The distribution and evolution of  $C_{\text{ant}}$  in the bottom layer of the SO are related to complex interactions between climatic forcing, air-sea  $CO_2$  exchange at formation sites, as well as biological and physical processes during AABWs circulation. The dataset that we collected regularly in the Enderby basin over the last 20 years (1998-2018) in the frame of the OISO project, together with historical observations obtained in 1978, 1985 and 1987 (GEOSECS and INDIGO cruises), allows the investigation of  $C_{\text{ant}}$  changes in AABW over 40 years in this region. The focus on the AABW variability is made by defining a Low Antarctic Bottom Water (LAABW) as described in the Section 2.3. Our results suggest that the accumulation of  $C_{\text{ant}}$  explains most, but not all, of the observed increase in  $C_T$ . We also detected a decrease in  $O_2$  that is consistent with the large-scale signal reported by Schmidtke et al. (2017), possibly due to a decrease in AABWs formation (Purkey and Johnson, 2012). Our data further indicate rapid anomalies in some periods suggesting that for decadal to long-term estimates care have to be taken when analyzing 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). Our results also show different  $C_{\text{ant}}$  trends on short periods, with a maximum increase of  $6.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  between 1987 and 2004 and an apparent stability in the last 20 years (despite an increase in  $C_T$ ). This suggests that AABWs have stored less  $C_{\text{ant}}$  in the last decade, but our understanding of the processes that explain this signal is not clear. This might be the result of the reduced  $CO_2$  uptake in the SO in the 1990s (Le Quéré et al., 2007; Landschützer et al., 2015), but this is not yet verified from direct  $C_T$  or  $fCO_2$  observations in AABW formation regions due to the lack of winter data and very large variability during summer. This calls for more data

collection and investigations in these regions. The apparent stability of  $C_{\text{ant}}$  in the LAABW since 1998 could also be directly linked to a decrease in AABWs formation in the 1990s (Purkey and Johnson, 2012) or a change in the contributions of AABWs from different sources, especially in the Prydz Bay region (Williams et al., 2016). In these scenarios, an increased contribution of  $C_T$ -rich and  $O_2$ -poor older LCDW along AABWs transit would also explain the decoupling between  $C_{\text{ant}}$  and  $C_T$  (increase in  $C_{\text{nat}}$ ) and decrease in  $O_2$  concentrations observed in recent years, even if we tried to isolate this specific feature in our data selection. The decoupling between  $C_{\text{ant}}$  and  $C_T$  is not a unique feature, as it was also reported along the SR03 section between Tasmania and Antarctica, most probably due to advection of  $C_T$ -rich waters (Pardo et al., 2017). This highlights the importance of the ocean circulation in influencing the temporal  $C_T$  and  $C_{\text{ant}}$  inventories changes (De Vries et al., 2017) and the need to better separate anthropogenic and natural variability based on time-series observations.

The evaluation and understanding of decadal  $C_{\text{ant}}$  changes in deep and bottom ocean waters are still challenging, as the  $C_{\text{ant}}$  concentrations remain low compared to  $C_T$  measurements accuracy (at best  $\pm 2 \mu\text{mol.kg}^{-1}$ , Bockmon and Dickson, 2015) and uncertainties of data-based methods ( $\pm 6 \mu\text{mol.kg}^{-1}$ ). Long-term repeated and qualified observations (at least 30 years) are needed to accurately detect and separate the anthropogenic signal from the internal ocean variability; we thus only start to document these trends that should now help to identify shortcomings in models regarding the carbon storage in the deep SO (e.g. Frölicher et al., 2014). As changes in the SO (including warming, freshening, oxygenation/deoxygenation,  $\text{CO}_2$  and acidification) are expected to accelerate in the future in response to anthropogenic forcing and climate change (e.g. Heuzé et al., 2014; Hauck et al., 2015; Ito et al., 2015, Yamamoto et al., 2015), it is important to maintain time-series observations to complement the GO-SHIP strategy, and to occupy more regularly other sectors of the SO (Rintoul et al., 2012). In this context, we hope to maintain our observations in the Southern Indian Ocean in the next decade, and with ongoing synthetic products activities such as GLODAPv2 (Olsen et al., 2016, 2019), SOCAT (Bakker et al., 2016) and more recently the SOCCOM project (Williams et al., 2018), to offer a solid database to validate ocean biogeochemical models and coupled climate/carbon models (Russell et al. 2018), and ultimately reduce uncertainties in future climate projections.

#### **Data availability**

GEOSECS, INDIGO and OISO 1998-2011 data are publicly available at the Ocean Carbon Data System (OCADS; [https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2\\_2019](https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2_2019)). OISO original data are available at: [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 GLODAPv2\_2021.

#### **Author contributions**

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.

#### **Competing interests**

The authors declare that they have no conflict of interest.

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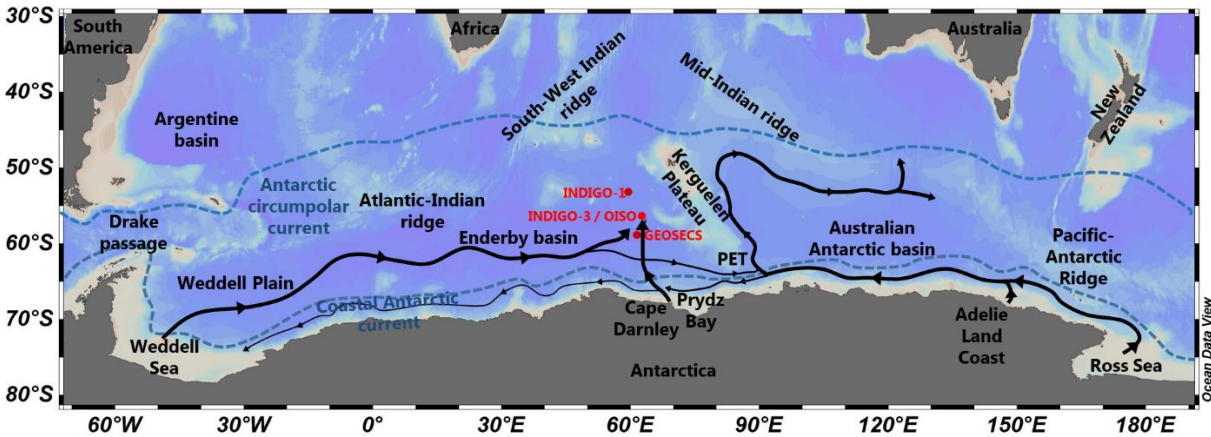
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Table 1. List of the cruises used in this study.

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

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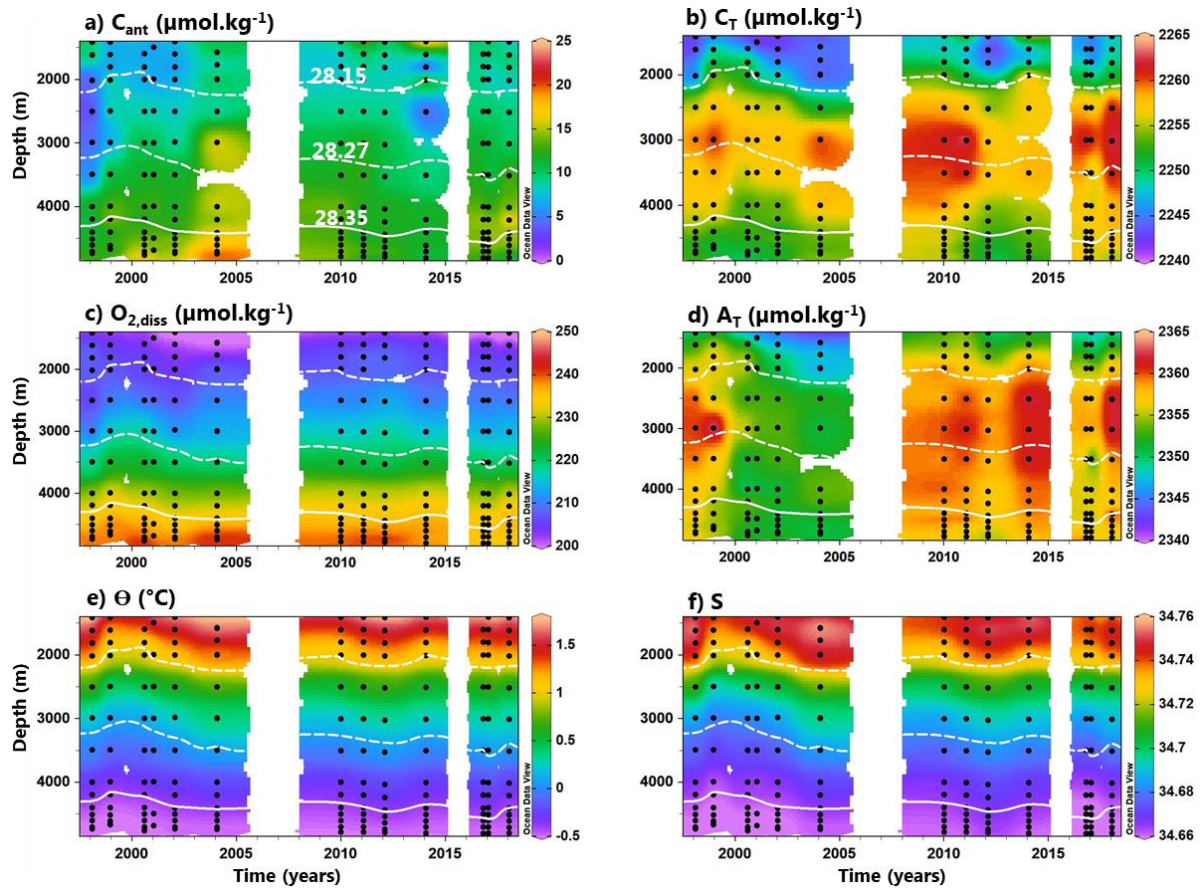
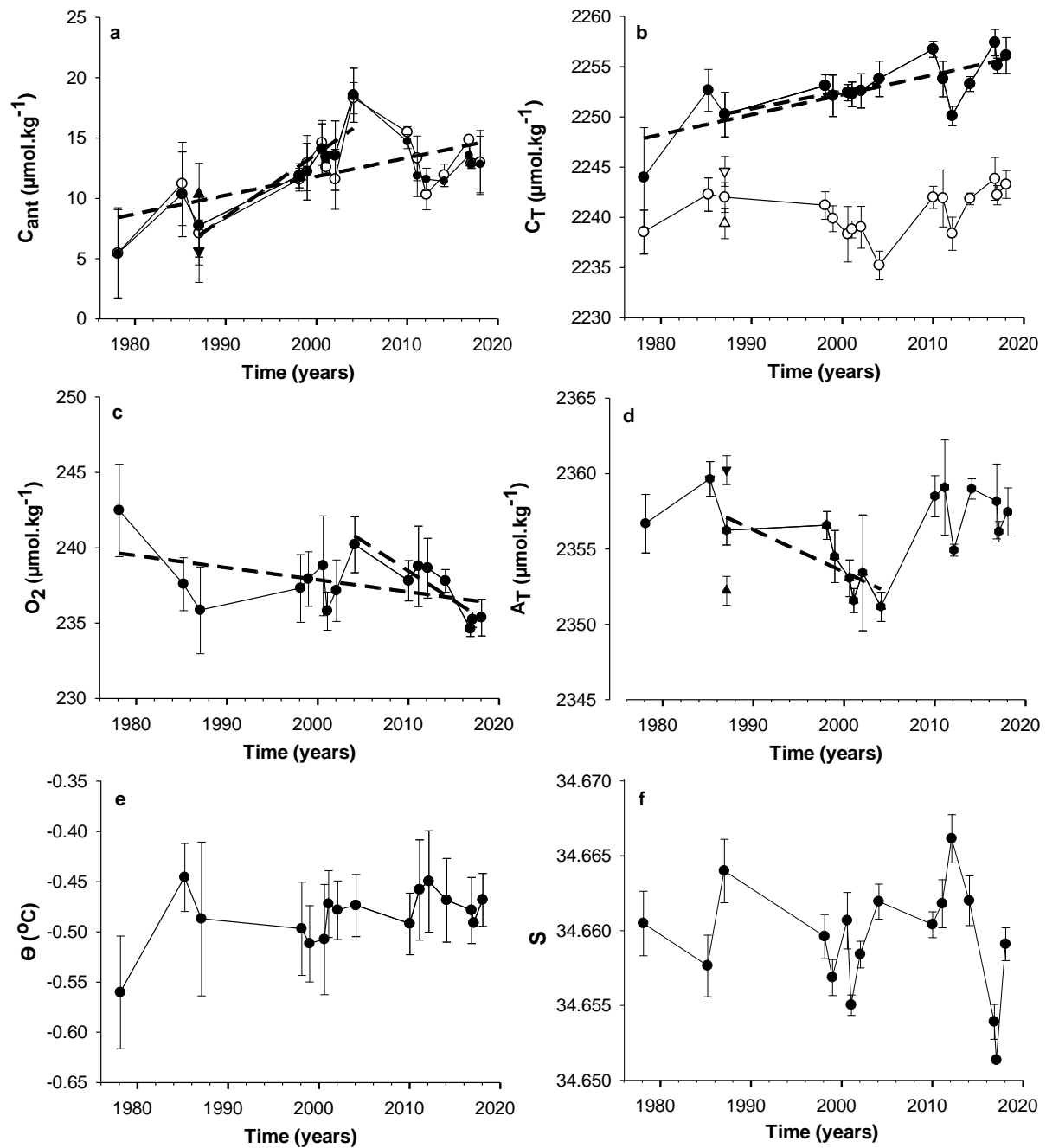


Figure 2. Hovmöller section of (a)  $C_{ant}$  via TrOCA, (b)  $C_T$ , (c)  $O_2$ , (d)  $A_T$ , (e)  $\theta$  and (f)  $S$  based on the OISO data presented in Table 1. Data points are represented by black dots. The white isolines represent the water masses separation by  $\gamma^n$  (from the bottom: LAABW, UAABW and LCDW). Figure produced with ODV (Schlitzer et al., 2019).

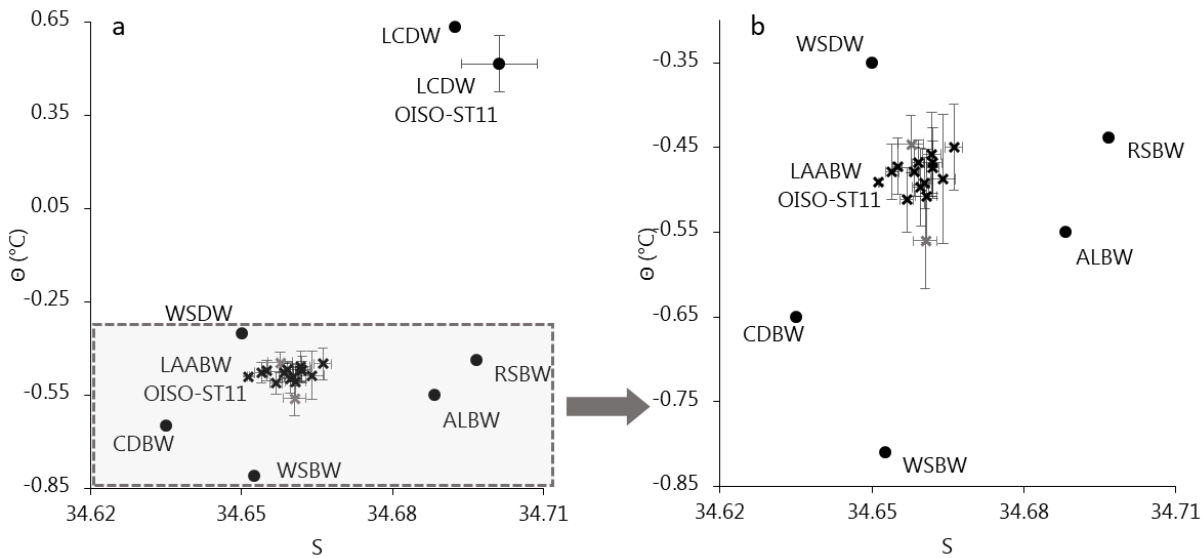


**Figure 3. Interannual variability (dash lines lines) and significant trends (at 95 %, see Table 2; dotted lines) for the 40 years of observation of the OISO-ST11 LAABW properties, including (a)  $C_{ant}$  by the TrOCA (black circles and triangles) and the  $C^0$  (open circles) method, (b)  $C_T$  (black circles) and  $C_{nat}$  (open circles), (c)  $O_2$ , (d)  $A_T$ , (e)  $\Theta$  and (f)  $S$ . For (a)  $C_{ant}$ , (b)  $C_{nat}$  and (d)  $A_T$ , the triangles pointing down and up correspond to INDIGO-3 value without and with  $-8 \mu\text{mol.kg}^{-1}$  of correction on the  $A_T$ , respectively (see Supp. Mat. for more details).**

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

Period	S	$\Theta$ °C	Si $\mu\text{mol.kg}^{-1}$	$\text{NO}_3$ $\mu\text{mol.kg}^{-1}$	$\text{O}_2$ $\mu\text{mol.kg}^{-1}$	$\text{A}_\text{T}$ $\mu\text{mol.kg}^{-1}$	$\text{C}_\text{T}$ $\mu\text{mol.kg}^{-1}$	$\text{C}_\text{ant}$ TrOCA $\mu\text{mol.kg}^{-1}$
1978-2018	-0.001 $\pm$ 0.001	0.01 $\pm$ 0.01	-1.2 $\pm$ 0.9	0.2 $\pm$ 0.2	<b>-0.8 <math>\pm</math> 0.4</b>	-0.1 $\pm$ 0.1	<b>2.0 <math>\pm</math> 0.5</b>	<b>1.4 <math>\pm</math> 0.5</b>
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	0.6 $\pm$ 0.1	<b>1.6 <math>\pm</math> 0.5</b>	1.1 $\pm$ 0.8
1987-2004	-0.003 $\pm$ 0.002	0.01 $\pm$ 0.01	<b>-6.5 <math>\pm</math> 1.8</b>	0.9 $\pm$ 0.9	1.7 $\pm$ 1.0	<b>-1.9 <math>\pm</math> 1.1</b>	<b>1.8 <math>\pm</math> 0.4</b>	<b>5.2 <math>\pm</math> 1.1</b>
2004-2018	-0.006 $\pm$ 0.003	0.01 $\pm$ 0.01	-1.8 $\pm$ 4.5	-0.5 $\pm$ 1.0	<b>-3.9 <math>\pm</math> 0.7</b>	3.4 $\pm$ 0.2	1.7 $\pm$ 1.9	-3.5 $\pm$ 1.5



**Figure 4. (a) Full  $\Theta$ -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 LAABW and OISO-ST11 LCDW. Error bars are calculated from the individual annual averaged values for the OISO-ST11 LAABW and from all data for the OISO-ST11 LCDW. For the OISO-ST11 LAABW, the grey cross are the GEOSECS (lowest  $\Theta$ ) and INDIGO-1 (highest  $\Theta$ ) values.**

**Table 3. Compilation of  $C_{\text{ant}}$  sequestration investigations in the AABWs ( $\gamma^n \geq 28.25 \text{ kg.m}^{-3}$ ) using the TrOCA method. The  $C_{\text{ant}}$  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 sink high salinity surface water. The mean values published by Roden et al. (2016) for the AABWs present WSDW characteristics but can be a mix of CDBW and LCDW.**

Source	Location	Water masses considered	Year	$C_{\text{ant}}$ $\mu\text{mol.kg}^{-1}$
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-LCDW- CDBW	2006	25
van Heuven et al. (2011) Fig.13	Weddell gyre (0°E; 55°-71°S)	WSBW	2005	16
This study	Enderby basin (56.5°S-63°E)	WSDW-CDBW- RSBW-ALBW	1978-1987	$8 \pm 3$
			1987-1998	$10 \pm 4$
			1987-2004	$13 \pm 4$
			1998-2004	$14 \pm 2$
			2010-2018	$13 \pm 1$
			<b>1978-2018</b>	<b><math>12 \pm 3</math></b>