



Carbon uptake and biogeochemical change in the Southern Ocean, south of Tasmania

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Abstract. Biogeochemical change in the water masses of the Southern Ocean, south of Tasmania, was assessed for the 16-year period between 1995 and 2011 using data from four summer repeats of the WOCE-JGOFS–CLIVAR–GO-SHIP (Key et al., 2015; Olsen et al., 2016) SR03 hydrographic section (at $\sim 140^\circ$ E). Changes in temperature, salinity, oxygen, and nutrients were used to disentangle the effect of solubility, biology, circulation and anthropogenic carbon (C_{ANT}) uptake on the variability of dissolved inorganic carbon (DIC) for eight water mass layers defined by neutral surfaces (γ^n). C_{ANT} was estimated using an improved back-calculation method. Warming ($\sim 0.0352 \pm 0.0170^\circ\text{C yr}^{-1}$) of Subtropical Central Water (STCW) and Antarctic Surface Water (AASW) layers decreased their gas solubility, and accordingly DIC concentrations increased less rapidly than expected from equilibration with rising atmospheric CO_2 ($\sim 0.86 \pm 0.16 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ versus $\sim 1 \pm 0.12 \mu\text{mol kg}^{-1} \text{yr}^{-1}$). An increase in apparent oxygen utilisation (AOU) occurred in these layers due to either remineralisation of organic matter or intensification of upwelling. The range of estimates for the increases in C_{ANT} were 0.71 ± 0.08 to $0.93 \pm 0.08 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ for STCW and 0.35 ± 0.14 to $0.65 \pm 0.21 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ for AASW, with the lower values in each water mass obtained by assigning all the AOU change to remineralisation. DIC increases in the Sub-Antarctic Mode Water (SAMW, $1.10 \pm 0.14 \mu\text{mol kg}^{-1} \text{yr}^{-1}$) and Antarctic Intermediate Water (AAIW, $0.40 \pm 0.15 \mu\text{mol kg}^{-1} \text{yr}^{-1}$) layers were similar to the calculated C_{ANT} trends. For SAMW, the C_{ANT} increase tracked rising atmospheric CO_2 . As a consequence of the general DIC increase, decreases in total pH (pH_T) and aragonite saturation (Ω_{Ar})

were found in most water masses, with the upper ocean and the SAMW layer presenting the largest trends for pH_T decrease ($\sim -0.0031 \pm 0.0004 \text{yr}^{-1}$). DIC increases in deep and bottom layers ($\sim 0.24 \pm 0.04 \mu\text{mol kg}^{-1} \text{yr}^{-1}$) resulted from the advection of old deep waters to re-supply increased upwelling, as corroborated by increasing silicate ($\sim 0.21 \pm 0.07 \mu\text{mol kg}^{-1} \text{yr}^{-1}$), which also reached the upper layers near the Antarctic Divergence ($\sim 0.36 \pm 0.06 \mu\text{mol kg}^{-1} \text{yr}^{-1}$) and was accompanied by an increase in salinity. The observed changes in DIC over the 16-year span caused a shoaling (~ 340 m) of the aragonite saturation depth (ASD, $\Omega_{\text{Ar}} = 1$) within Upper Circumpolar Deep Water that followed the upwelling path of this layer. From all our results, we conclude a scenario of increased transport of deep waters into the section and enhanced upwelling at high latitudes for the period between 1995 and 2011 linked to strong westerly winds. Although enhanced upwelling lowered the capacity of the AASW layer to uptake atmospheric CO_2 , it did not limit that of the newly forming SAMW and AAIW, which exhibited C_{ANT} storage rates ($\sim 0.41 \pm 0.20 \text{mol m}^{-2} \text{yr}^{-1}$) twice that of the upper layers.

1 Introduction

The Southern Ocean is a key region in terms of climate change and climate variability, influencing the Meridional Overturning Circulation (MOC) and therefore modulating the global circulation and oceanic biogeochemical cycles (Sarmiento et al., 1998, 2004; Orr et al., 2005). Deep waters, formed in the North Atlantic, spread south and enter the Southern Ocean, where they mix with deep layers of the

Antarctic Circumpolar Current (ACC) and ultimately upwell between the Southern ACC Front and the Polar Front. The upwelled waters are eventually transformed into bottom, intermediate and mode waters, which are exported from the Southern Ocean to ventilate the thermocline and bottom layers of the major ocean basins. Some of the Southern Ocean waters subducted into the ocean interior return to the North Atlantic to balance the southward flux of North Atlantic Deep Water (Speer et al., 2000; Lumpkin and Speer, 2007; Iudicone et al., 2008).

Within the eastward flow of the ACC, major water exchange between the three ocean basins takes place. The circumpolar path of the ACC consists of various narrow jets associated with sharp fronts that separate waters with different characteristics (Orsi et al., 1995; Belkin and Gordon, 1996). These jets can reach deep layers and often meander, intensify, merge and split, conditioned by the topography of the ocean floor, the stratification of the ACC, and atmospheric variability (Moore et al., 1999; Sokolov and Rintoul, 2002, 2009; Peña-Molino et al., 2014). Movements in the jets enhance cross-stream transports and mesoscale activity that can result in local changes in water mass properties and this may complicate the computation of long-term changes in water mass properties (Rintoul and Bullister, 1999; Sallée et al., 2008; Peña-Molino et al., 2014).

Water mass formation and ventilation transport heat, salt, and dissolved gases from the atmosphere to the ocean interior and other basins (Sarmiento et al., 2004), with the Southern Ocean contributing $\sim 40\%$ to the anthropogenic CO_2 (C_{ANT}) inventory of the ocean (Sabine et al., 2004; Gruber et al., 2009; Khatiwala et al., 2009). Circulation and biological processes drive the redistribution of dissolved inorganic carbon (DIC) that ultimately affects the capacity of the waters to uptake more CO_2 . The uptake of CO_2 by the Southern Ocean presents strong spatiotemporal variability (Lenton et al., 2013), and this can lead to conflicting results for observational studies, models and atmospheric inversions, depending on the methodology used (Verdy et al., 2007; Lenton et al., 2012; Fay et al., 2014). Quantifying long-term changes in the carbon system is difficult due to the scarcity of data (Lenton et al., 2012; Kouketsu and Murata, 2014; Fay et al., 2014) and the influence of biological processes. Notably, long-term trends in C_{ANT} concentration are difficult to estimate due to its small signal ($\sim 3\%$) with respect to that of DIC in the ocean.

Once CO_2 dissolves in the ocean (DIC) it begins the process of ocean acidification, i.e. decreases the pH and the saturation state of calcium carbonate (CaCO_3) minerals such as calcite and aragonite (Feely et al., 2004; Bates et al., 2014), with potential to disrupt ecosystems and biological processes (Doney et al., 2009).

Numerous studies have documented warming and freshening of deep and bottom layers of the Southern Ocean in recent decades (see reviews by Jacobs, 2006 and van Wijk and Rintoul, 2014). The abyssal waters of the Australian–

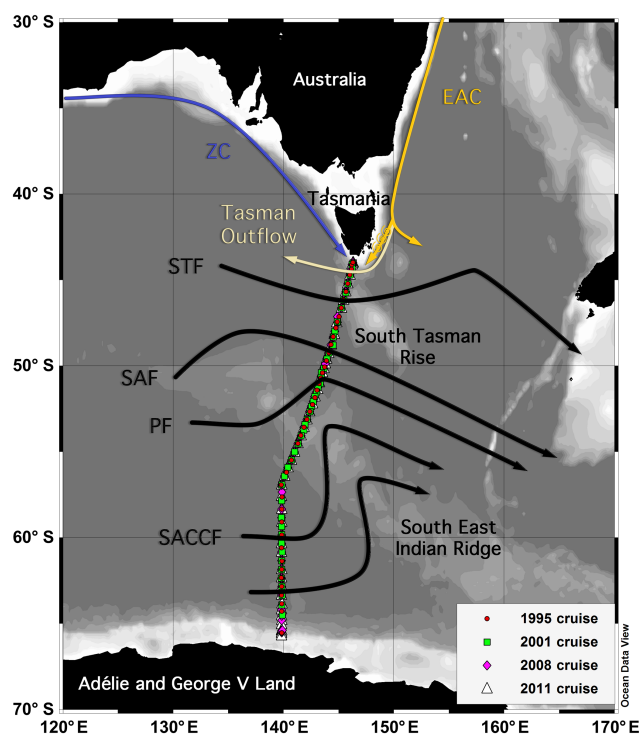


Figure 1. Transects of the four summer repeats of the G0-SHIP hydrodynamic line SR03 in the Southern Ocean south of Tasmania for the period 1995–2011 and the main hydrography features of the region. ZC = Zeehan Current. EAC = East Australian Current. Black arrows indicate the flow of the Antarctic Circumpolar Current (ACC), with STF = Subtropical Front; SAF = Sub-Antarctic Front, PF = Polar Front and SACCF = Southern ACC Front.

Antarctic Basin (A–AB) show the greatest freshening in the last 40 years (van Wijk and Rintoul, 2014). This freshening was accompanied by a warming of the deep–bottom layers, leading to a contraction in bottom waters by more than half their volume in the basin (Purkey and Johnson, 2012; van Wijk and Rintoul, 2014). Subsurface to intermediate layers have also warmed and freshened south of Australia (Bindoff and Church, 1992; Wong et al., 1999; Aoki et al., 2005). The solubility of gases and their distribution in the ocean depend on the dynamics and properties of water masses, and the thermohaline changes that have occurred south of Tasmania (Fig. 1) have implications in the carbon system of the region.

A reduction of the carbon sink of the Southern Ocean was observed between the 1980s and the early 2000s (Le Quéré et al., 2007; Lovenduski et al., 2008), with more recent studies suggesting a recovery and even intensification of the CO_2 uptake by 2011–2012 (Zickfeld et al., 2008; Fay et al., 2014; Landschützer et al., 2015). DeVries et al. (2017), used a global inverse model to postulate that changes in circulation are responsible for most of the variability in the oceanic CO_2 uptake, with the weakening of the upper-ocean circulation being responsible for the increase in oceanic carbon uptake

Table 1. Repeats of the GO-SHIP SR03 line on board the Aurora Australis from 1995 to 2011 with the expocode from GLODAPv2 database.

EXPOCODE	Dates (dd/mm/yyyy)	REF.
09AR19941213	20/12/1994–1/02/1995	1995
09AR20011029	29/10/2001–11/12/2001	2001
09AR20080322	23/03/2008–15/04/2008	2008
09AR20110124	4/01/2011–31/01/2011	2011

over the past decade. Ocean acidification has been observed in the whole Southern Ocean (Lauvset et al., 2015) and locally in the Atlantic and Pacific sectors (Williams et al., 2015; Hauri et al., 2015). South of Tasmania, McNeil et al. (2001) reported an increase in C_{ANT} uptake between 1968 and 1996 for the region 45–50° S. These authors reported, for the first time, C_{ANT} accumulation in the AABW and highlighted the importance of the formation of bottom and mode waters as a mechanism for transporting C_{ANT} to the ocean. In terms of ocean acidification, we are not aware of any study about trends in ocean acidification in the water masses south of Australia.

Considering the lack of observational estimates for recent biogeochemical changes in the A–AB as well as the large changes in CO_2 uptake and storage suggested by recent atmospheric and surface observations in the Southern Ocean (e.g. Fay et al., 2014; Landschützer et al., 2015) there is a need to provide a full ocean depth observational perspective on how the ocean is changing. The aim of this paper is to provide the first estimates of biogeochemical change in the water masses south of Tasmania, for the period 1995–2011, disentangling the effects that solubility, circulation, biology and C_{ANT} uptake have on the variability of DIC. We use data from four summer repeats of the (Key et al., 2015; Olsen et al., 2016) WOCE–JGOFS–CLIVAR–GO-SHIP hydrographic section SR03 (Fig. 1; Table 1), one of the most revisited sections in the Southern Ocean. Trends in oxygen (O_2), nutrients, and the carbon system parameters (i.e. DIC, total alkalinity (TA), anthropogenic carbon (C_{ANT}), total pH (pH_T) and percentage aragonite saturation (Ω_{Ar}) were estimated for the period 1995–2011, when both DIC and TA measurements are available. C_{ANT} estimates were obtained with a back-calculation method (Pardo et al., 2014). The changes were evaluated in the different water mass layers of the section defined by neutral surfaces (γ^n , McDougall et al., 1987).

2 Hydrography of the region

The dynamical structure of the region south of Tasmania (Fig. 1) is characterized by a number of fronts that separate the major water masses of the region (Sokolov and Rintoul, 2002, 2007, 2009). At the northern end of the section, the

presence of the weak Subtropical Front (STF, Fig. 1) separates warm, salty subtropical surface waters from cooler and fresher sub-Antarctic surface waters (Deacon, 1937). The northern end of the section is a complex mixing zone where waters transported down the east coast of Tasmania in a series of mesoscale eddies from the East Australian Current (EAC, Fig. 1) mix into the Subantarctic Zone, and also meet Zeehan Current (ZC, Fig. 1) waters transported down the west coast of Tasmania (Boland and Church, 1981; Baines et al., 1983; Speich et al., 2002; Davis, 2005; Ridgway et al., 2007; Sloyan et al., 2016). The EAC transported south of Tasmania forms a zonal jet towards the south-east Indian Ocean, known as the Tasman Outflow, that reaches the bottom of the Tasman slope and that is maintained all year round (Rintoul and Bullister, 1999; Ridgway et al., 2007). The encounter between these currents presents high variability at the northern end of the section (Fig. 1).

Farther south (Fig. 1), the Sub-Antarctic Front (SAF) and the Polar Front (PF) are regions of maximum transport in the ACC (Rintoul and Bullister, 1999; Sokolov and Rintoul, 2002). North of the SAF, deep winter convection generates Sub-Antarctic Mode Water (SAMW), a relatively uniform water mass that occupies subsurface layers down to ~ 600 m deep (McCartney, 1977; Rintoul and Bullister, 1999). SAMW constitutes the main part of the upper limb of the MOC, ventilating the thermocline of all the ocean basins (e.g. Speer et al., 2000; Sloyan and Rintoul, 2001). The SAF coincides with the deepening of the salinity minimum at intermediate depths (Whitworth III and Nowlin Jr., 1987), which is the signature of the Antarctic Intermediate Water (AAIW). The AAIW underlies the SAMW and also ventilates the global thermocline layers of the ocean. It is mainly formed in the south-east Pacific and it is continuously transformed on its way to the region south of Tasmania (Hanawa and Talley, 2001). South of the SAF, colder and fresher Antarctic Surface Water (AASW) covers the surface ocean. AASW originates from progressive warming of Winter Water (WW, Mosby, 1934), which can be perceived even in summer as a remnant layer of cold water at the base of the AASW (Rintoul et al., 1997).

The Southern ACC Front (SACCF, Fig. 1) is a deep front located south of the PF (Orsi et al., 1995) and can coincide with the southern boundary of the ACC, which is represented by the southern limit of the oxygen minimum (Orsi et al., 1995). The oxygen minimum is related to the advection of Upper Circumpolar Deep Water (UCDW) that originates in the Indian and Pacific Oceans (Callahan, 1972), from where it spreads south, mixes with deep layers of the ACC and ultimately upwells near the Antarctic continent as part of the lower cell of the MOC. Lower Circumpolar Deep Water (LCDW) is below UCDW and is identified by a salinity maximum and contributes to upwelling around Antarctica. The precursor of the LCDW is North Atlantic Deep Water (NADW), which originates in the Labrador and Nordic seas of the North Atlantic polar region (Dickson and Brown,

1984) that flows southward to enter the ACC as part of the MOC (Callahan, 1972; Orsi et al., 1995; Johnson, 2008).

At the southern end of the section, the Antarctic Slope Front forms the boundary between cold and fresh shelf water and relatively warm and salty waters offshore (Jacobs, 1991). Polynyas along the Adélie and George V Land coast (Fig. 1) contribute to the formation of Adélie Land Bottom Water (ALBW), which is a mixture of High-Salinity Shelf Water (HSSW) resulting from brine rejection during ice formation and ultra-modified LCDW (Foster and Carmack, 1976; Rintoul, 1998; Marsland et al., 2004). ALBW constitutes $\sim 25\%$ of the total volume of water $< 0^\circ\text{C}$ in the ocean (Rintoul, 1998). The bottom waters near the southern end of the section also contain a component of Ross Sea Bottom Water (RSBW) that originates to the east and is deflected westwards towards the A–AB as it is transported down the continental slope. The RSBW is modified when it arrives at the location of the SR03 section by mixing with deep layers of the ACC and recently formed ALBW (Gordon and Tchernia, 1972; Rintoul, 1998). The ALBW and the modified RSBW together ventilate the abyssal layers of the A–AB before spreading north to ventilate the Indian and Pacific basins (Mantyla and Reid, 1995; Fukamachi et al., 2010).

3 Data and method

3.1 Data

The SR03 hydrographic section between Tasmania and Antarctica (Fig. 1) was occupied between 1991 and 2011. Measurements of total alkalinity (TA) were only available from the beginning of 1995, and our evaluation of biogeochemical changes is limited to four summer sections occupied for the period 1995–2011 (Table 1). A winter cruise in 1996 was not considered in this study in order to minimise seasonal biases.

Water column salinity, temperature, pressure and dissolved oxygen (O_2) were collected from the conductivity–temperature–depth (CTD) device with an accuracy of ± 0.002 for salinity and temperature, $\pm 0.015\%$ of full-scale range for pressure and $\pm 1\%$ for O_2 , according to WOCE standards (Joyce and Corry, 1994). Samples from the Niskin bottles were analysed for DIC by coulometry and TA by open cell potentiometric titration (Dickson et al., 2007). Certified reference material provided by A. Dickson, Scripps Institution of Oceanography, were used as reference standards for DIC and TA. The precisions of DIC and TA measurements improved slightly on more recent sections, and for all sections were better than $\pm 2\mu\text{mol kg}^{-1}$, for both variables, based on analysis of duplicate samples and certified reference material. Samples for dissolved O_2 were measured using modified Winkler titrations (Hood et al. 2010), with an estimated accuracy and precision of $\pm 0.3\%$ for the sections. For the 1995 cruise, sensor-based O_2 was used instead of

sampled O_2 because of the poor quality of many of the Winkler measurements.

Other variables of the dissolved CO_2 system were calculated from the DIC and TA measurements. We calculate pH_T and Ω_Ar (from measured DIC and TA) using the CO2sys program from Lewis and Wallace (1998) adapted to MATLAB by van Heuven et al. (2011). We use the constants for the carbonic acid from Mehrbach et al. (1973) refit by Dickson and Millero (1987), the CO_2 solubility equation from Weiss (1974), dissociation constants for sulfate from Dickson (1990) and borate constant from Uppstrom (1974). Aragonite saturation states (Ω_Ar) are calculated because it is a less stable form of CaCO_3 than calcite and is the predominant biogenic form of CaCO_3 precipitated by calcifying organisms.

Data presented here were interpolated to a regular grid, and the water mass layers were defined as layers between neutral surfaces (γ^n) determined using potential temperature and salinity and published literature values (Table 2, Sect. 2). The first 50 m of the water column were eliminated in order to reduce the short-timescale variability in surface properties. The use of γ^n to identify the water mass layers reduces the variability due to isopycnal heave caused for example by eddies and internal waves (McDougall et al., 1987; Bindoff and McDougall, 1994; Jackett and McDougall, 1997). The upper ocean layers south of the SAF were divided into the AASW layer to the north of the PF, and the AASW_{upw} layer that is composed of surface waters south of the PF (Fig. 1). ADLBW and RSBW were included in the AABW layer (Table 2). The ACC fronts along the SR03 section (Table 3) were defined as a function of hydrographic variables following Sokolov and Rintoul (2002).

3.2 Estimates of anthropogenic carbon (C_ANT)

C_ANT was estimated using a back-calculation method (Chen and Millero, 1979; Gruber, 1998) combined with an optimum multi-parameter (OMP) analysis (Tomczak, 1981), described by Pardo et al. (2014). This technique has the advantage of considering water mass mixing and the temporal variability of the air–sea CO_2 disequilibrium. The accuracy of the method is $\pm 6\mu\text{mol kg}^{-1}$ (Pardo et al., 2014). Back-calculation methods assume the ocean is at steady state for dynamical and biological processes and estimate C_ANT ($C_\text{ANT_BC}$) as an excess of DIC in the ocean resulting from the increase in atmospheric CO_2 due to anthropogenic emissions as follows:

$$C_\text{ANT_BC} = \text{DIC} - \text{DIC}^\text{BIO} - \text{DIC}^\pi, \quad (1)$$

where DIC^BIO is the biological contribution to DIC, and DIC^π is the preformed concentration of DIC in pre-industrial times.

Table 2. Definition of the water mass layers between neutral surfaces (γ^n) and references used to accordingly decide the limits of the layers. We also consider limits in salinity, depth and position of the ACC fronts (see Table 3) to differentiate better some of the layers.

Layer	Definition	Complete name	Reference
STCW	$\gamma^n < 26.85$ & $S \geq 34.3$	Subtropical Central Water	Rintoul (1998)
AASW	$\gamma^n < 27.7$ & $S < 34.3$	Antarctic Surface Water	Rintoul (1998); Williams et al. (2015)
AASW _{upw}	$\gamma^n \geq 27.7$ & depth ≤ 300 m	Antarctic Surface Water	–
SAMW	$26.85 \leq \gamma^n < 27.2$ & $S \geq 34.3$	Sub-Antarctic Mode Water	Rintoul (1998); Rintoul and Bullister (1999)
AAIW	$27.2 \leq \gamma^n < 27.7$ & $S \geq 34.3$	Antarctic Intermediate Water	Rintoul and Bullister (1999)
UCDW	$27.7 \leq \gamma^n < 28.18$ & depth > 300 m	Upper Circumpolar Deep water	Williams et al. (2015)
LCDW	$28.18 \leq \gamma^n < 28.25$	Lower Circumpolar Deep Water	Lacarra et al. (2011); Williams et al. (2015)
AABW	$\gamma^n \geq 28.25$	Antarctic Bottom Water	Rintoul and Bullister (1999); Williams et al. (2015)

The term DIC^{BIO} is calculated (Chen et al., 1982; Ikegami and Kanamori, 1983) by the following:

$$\text{DIC}^{\text{BIO}} = \frac{\text{AOU}}{R_C} + \frac{1}{2} \left[\text{TA} - \text{TA}^0 + \text{AOU} \cdot \left(\frac{1}{R_N} + \frac{1}{R_P} \right) \right], \quad (2)$$

where R_C , R_N and R_P are the stoichiometric ratios of carbon, nitrate and phosphate respectively, referred to O_2 consumption by respiration–remineralisation processes that are considered constant (1.45, 9 and 125, respectively, Broecker, 1974; Anderson and Sarmiento, 1994; Martiny et al., 2013).

The term $\frac{\text{AOU}}{R_C}$ represents the remineralisation of organic matter, with the apparent oxygen utilisation (AOU) defined as $\text{AOU} = \text{O}_{\text{SAT}} - \text{O}_2$, i.e. the difference between the saturation of oxygen (O_{SAT}) at the potential temperature (θ) and salinity of the measured O_2 , and the measured O_2 . The term $\frac{1}{2} \left[\text{TA} - \text{TA}^0 + \text{AOU} \cdot \left(\frac{1}{R_N} + \frac{1}{R_P} \right) \right]$ represents the dissolution or precipitation of CaCO_3 , with TA^0 , the preformed alkalinity, obtained in the water formation sites using regional parameterisations as functions of salinity, θ and phosphate (Pardo et al., 2011; Vázquez-Rodríguez et al., 2012; Supplement Sect. S3, Table S3).

The preformed pre-industrial term, DIC^π , is the total concentration of carbon dioxide in seawater saturated with respect to the pre-industrial $p\text{CO}_2$ ($\text{DIC}_{\text{SAT}}^\pi$) and corrected for an air–sea CO_2 disequilibrium (CDIS^π) term:

$$\text{DIC}^\pi = \text{DIC}_{\text{SAT}}^\pi + \text{CDIS}^\pi. \quad (3)$$

CDIS^π is time dependent:

$$\text{CDIS}^\pi = \text{CDIS} - \delta\text{CDIS}, \quad (4)$$

where CDIS is the current disequilibrium between the ocean and the atmosphere $p\text{CO}_2$, and δCDIS is the change in the disequilibrium from pre-industrial to current times. CDIS was obtained by similar parameterisations to those used for TA^0 , combined with monthly mean values of atmospheric

CO_2 values from the NOAA network (Dlugokencky et al., 2016) (see Sect. S3 and Table S3). The δCDIS values were obtained from results of the $1/10^\circ$ resolution carbon model OFAM3-WOMBAT (Sect. S1).

Interior values of preformed variables (TA^0 and $\text{DIC}_{\text{SAT}}^\pi$) and CDIS^π were obtained using an OMP analysis to mix endmembers as described in Sects. S2 and S3, using the endmembers in Table S1 in the Supplement. The OMP analysis is based on the assumption that a property measured in a certain point is the result of linear mixing between endmembers, known as source water types (SWTs). A system of equations is created for each measurement point and is solved to obtain the fractions of the different SWTs (Sect. S2). The application of the OMP analysis requires good regional hydrodynamic knowledge as the results are strongly dependent on the definition of the SWTs (Tomczak, 1981). We used 11 SWTs to characterise the biogeochemical properties of the waters in the SR03 section and the SWT properties were assumed to be constant with time (Table S1, Fig. S1).

Small negative values of $\text{C}_{\text{ANT_BC}}$ can occur due to an overestimation of the CDIS^π term (Eq. 4), which acts as measure of the age of the water mass and has high values in old deep layers (see Table S1). These negative values of $\text{C}_{\text{ANT_BC}}$ were found at some points in deep waters of the section (mainly UCDW and LCDW layers) and were small (between 0 and $-2 \mu\text{mol kg}^{-1}$, i.e. less than the accuracy of the methodology) and changed to zero for our analysis.

3.3 Changes in the carbon system

Changes in carbon system parameters $\left(\frac{\partial \text{DIC}}{\partial t}, \frac{\partial \text{TA}}{\partial t}, \frac{\partial \text{C}_{\text{ANT_BC}}}{\partial t}, \frac{\partial \text{pH}}{\partial t} \text{ and } \frac{\partial \Omega_{\text{Ar}}}{\partial t} \right)$ were estimated using linear regressions with time for the period 1995–2011 in each one of the water mass layers defined by their γ^n condition (Table 2). The trends were estimated using all the points in each water mass layer, and only those linear trends with $p < 0.05$ and $r \geq 0.2$ were considered statistically

significant for discussion. We show the value of the root mean square error (RMSE or square root of the variance of the residuals), which can be interpreted in large part as unexplained variance caused by short-timescale processes and the different seasonal timings of the cruises. RMSE has the same units as the response variable.

With respect to the total change in DIC $\left(\frac{\partial \text{DIC}}{\partial t}\right)$, our goal is to disentangle the effects that solubility, circulation, biology and C_{ANT} uptake have on the variability of DIC. The total change in DIC $\left(\frac{\partial \text{DIC}}{\partial t}\right)$ in a water mass is due to changes in the atmosphere–ocean interchange, biological processes and circulation processes. In order to account for the change in DIC due the atmosphere–ocean interchange and biological processes, we compare $\frac{\partial \text{DIC}}{\partial t}$ to $\frac{\partial C_{\text{ANT_BC}}}{\partial t}$ and $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$, Eq. (2). The change in DIC not explained by $\frac{\partial C_{\text{ANT_BC}}}{\partial t}$ or $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$ will then be due to circulation processes.

In order to compare $\frac{\partial \text{DIC}}{\partial t}$ to $\frac{\partial C_{\text{ANT_BC}}}{\partial t}$ we need to consider how the change in $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$ and $\text{DIC}^{\pi} \left(\frac{\partial \text{DIC}^{\pi}}{\partial t}\right)$ (terms of Eq. 1, Sect. 3.2) affect the changes in $C_{\text{ANT_BC}}$.

The term $\frac{\partial \text{DIC}^{\pi}}{\partial t}$ can be expressed as follows:

$$\frac{\partial \text{DIC}^{\pi}}{\partial t} = \frac{\partial \text{DIC}_{\text{SAT}}^{\pi}}{\partial t} - \frac{\partial \text{CDIS}^{\pi}}{\partial t}. \quad (5)$$

The terms $\frac{\partial \text{CDIS}^{\pi}}{\partial t}$ and $\frac{\partial \text{DIC}_{\text{SAT}}^{\pi}}{\partial t}$ reflect changes in the properties of the water masses over time, primarily temperature and salinity change due to mixing and heating or cooling. CDIS^{π} and $\text{DIC}_{\text{SAT}}^{\pi}$ are defined at the ocean surface (in each of the SWTs: Table S1) and are calculated at each point in the ocean interior using the OMP analysis (Sects. S2 and S3). Because a change in temperature and/or salinity in the water is solved by the OMP analysis as a change in the SWTs fractions, this also produces varying CDIS^{π} and $\text{DIC}_{\text{SAT}}^{\pi}$. No significant trends were obtained for CDIS^{π} in any of the layers. $C_{\text{ANT_BC}}$ (Eq. 1) is not affected by the changes in solubility occurring from one voyage to another (thus neither is $\frac{\partial C_{\text{ANT_BC}}}{\partial t}$), since any change in temperature or salinity is cancelled out by the subtraction of DIC^{π} with respect to DIC in Eq. (1) (Sect. 3.2) and by O_2 respect to O_{SAT} in the DIC^{BIO} term (Eq. 2). However, $\frac{\partial \text{DIC}}{\partial t}$ based on the measured DIC in the sections will be affected by changes in the solubility over time, and this difference needs to be accounted for when comparing $\frac{\partial \text{DIC}}{\partial t}$ with $\frac{\partial C_{\text{ANT_BC}}}{\partial t}$ to obtain a better approximation for the change in C_{ANT} : $\frac{\partial C_{\text{ANT}}}{\partial t} = \frac{\partial C_{\text{ANT_BC}}}{\partial t} + \frac{\partial \text{DIC}^{\pi}}{\partial t}$.

The term $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$ can be influenced by changes with time of alkalinity due to changes in the rate of carbonate precipitation–dissolution and of AOU due to changes in the rate of remineralisation and in circulation. In the present study only surface waters of the SR03 section present changes DIC^{BIO} between 1995 and 2011. Numerous studies have reported a strong influence of biological communities in the seasonal cycle of dissolved O_2 in surface waters (Bender

Table 3. Location of the ACC fronts south of Tasmania for each of the cruises following the definitions of Sokolov and Rintoul (2002) for hydrographic data. * The range in the location of the SAF for the 2011 cruise could be related to a diversification of the PF or SAF into different jets (Sokolov and Rintoul, 2009) but also to crossing the meander of the front twice.

Cruise	STF	SAF	PF	SACCF
1995	~ 45.4° S	~ 51.8° S	~ 55.2° S	~ 63.8° S
2001	~ 46.4° S	~ 50.1° S	~ 54.5° S	~ 63.7° S
2008	~ 46.4° S	~ 52.2° S	~ 54.6° S	~ 63.5° S
2011	~ 45° S	~ 52.16–50.5° S*	~ 54.5° S	~ 63.4° S

et al., 1996; Moore and Abbott, 2000; Sambrotto and Mace, 2000; Trull et al., 2001a). Inter-annual variability in O_2 in upper layers of the Southern Ocean have also been related to changes in the entrainment of deeper waters into the mixed layer due to the mixed layer depth variability (Matear et al., 2000; Verdy et al., 2007; Sabine et al., 2008; Sallée et al., 2012). Although some studies found long-term decreases in O_2 due to circulation in deep waters of the Weddell Sea (van Heuven et al., 2014) and for the first 1000 m of the global ocean (Helm et al., 2011), significant long-term trends in O_2 due to circulation and remineralisation processes have not yet been reported for surface waters of the Southern Ocean. Thus, the term $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$ may also contribute to variation in $\frac{\partial C_{\text{ANT_BC}}}{\partial t}$, since part of the changes in AOU with time reflect changes in circulation that we cannot separate from those in remineralisation. We consider the best approximation for the change in C_{ANT} as a range depending on the possible effect of biology and circulation processes on $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$. If the value of $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$ is due to the variability in the remineralisation rates and the change in solubility is considered, the estimate $\frac{\partial C_{\text{ANT_BC}}}{\partial t}$ will be the lower limit of the range (lower limit of $\frac{\partial C_{\text{ANT}}}{\partial t} = \frac{\partial C_{\text{ANT_BC}}}{\partial t} + \frac{\partial \text{DIC}^{\pi}}{\partial t}$). For the upper limit of the range, we consider that the value of $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$ is due to changes in circulation and the upper limit of the range is obtained by $\frac{\partial C_{\text{ANT}}}{\partial t} = \frac{\partial C_{\text{ANT_BC}}}{\partial t} + \frac{\partial \text{DIC}^{\pi}}{\partial t} + \frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$. We assume that the changes in DIC^{BIO} due to circulation do not affect the amount of DIC in the layer. This assumption is one of the caveats of the methodology, since we cannot know how much of the change in DIC is associated with changes in circulation, i.e. how much of the change in DIC is a change in non-anthropogenic DIC. We will discuss this more in Sect. 6.2.

4 Results

The different trends in biogeochemical properties are summarised in Table 4. The biogeochemical changes between 1995 and 2011 are presented for each of the water mass layers and the effect of changes in solubility, biological processes and circulation in the estimates of $\frac{\partial C_{\text{ANT}}}{\partial t}$ and $\frac{\partial \text{DIC}}{\partial t}$.

Table 4. Trends in the water mass layers for the period 1995–2011 of dissolved inorganic carbon ($\partial \text{DIC} / \partial t$), anthropogenic carbon ($\partial \text{C}_{\text{ANT}} / \partial t$), silicate ($\partial \text{SiO}_4 / \partial t$) total pH ($\partial \text{pH}_T / \partial t$), aragonite saturation ($\partial \Omega_{\text{Ar}} / \partial t$) and percentage of change in the aragonite saturation ($\% \partial \Omega_{\text{Ar}} / \partial t$). RMSE = root mean square error. * Trends in C_{ANT} for the AASW and AASW_{upw} layers considering an approximate value for the increase in DIC due to the advection of old deep waters to the section (see Sect. 4.4.1 in the text). ** The value of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$ in the AABW layer is considered negligible because it falls below the accuracy of the back-calculation method.

Layer	$\partial \text{DIC} / \partial t$ ($\mu\text{mol kg}^{-1} \text{ yr}^{-1}$)	$\partial \text{C}_{\text{ANT}} / \partial t$ ($\mu\text{mol kg}^{-1} \text{ yr}^{-1}$)	$\partial \text{SiO}_4 / \partial t$ ($\mu\text{mol kg}^{-1} \text{ yr}^{-1}$)	$\partial \text{pH}_T / \partial t$ (yr^{-1})	$\partial \Omega_{\text{Ar}} / \partial t$ (yr^{-1})	$\%$ $\partial \Omega_{\text{Ar}} / \partial t$ ($\% \text{ yr}^{-1}$)
	1995–2011	1995–2011	1995–2011	1995–2011	1995–2011	1995–2011
STCW	0.86 ± 0.07 , RMSE = 7	$[0.71 \pm 0.08 - 0.93 \pm 0.08]$	–	-0.0027 ± 0.0001 , RMSE = 0.01	-0.009 ± 0.001 , RMSE = 0.15	-0.42 ± 0.17
AASW	0.85 ± 0.14 , RMSE = 21	$[0.35 \pm 0.14 - 0.85 \pm 0.21]$ * $[0.35 \pm 0.14 - 0.65 \pm 0.21]$	–	-0.0035 ± 0.0002 , RMSE = 0.03	-0.009 ± 0.001 , RMSE = 0.14	-0.61 ± 0.19
AASW _{upw}	0.61 ± 0.10 , RMSE = 9	$[0 - 0.41 \pm 0.16]$ * $[0 - 0.21 \pm 0.16]$	0.36 ± 0.06 , RMSE = 6	-0.0015 ± 0.0004 , RMSE = 0.04	–	–
SAMW	1.10 ± 0.14 , RMSE = 16	0.92 ± 0.09 , RMSE = 10	–	-0.0031 ± 0.0003 , RMSE = 0.03	-0.011 ± 0.001 , RMSE = 0.17	-0.67 ± 0.20
AAIW	0.40 ± 0.15 , RMSE = 24	0.42 ± 0.06 , RMSE = 10	–	-0.0017 ± 0.0002 , RMSE = 0.03	–	–
UCDW	0.29 ± 0.02 , RMSE = 5	–	0.22 ± 0.04 , RMSE = 12	-0.0012 ± 0.0001 , RMSE = 0.03	–	–
LCDW	0.20 ± 0.02 , RMSE = 4	–	0.27 ± 0.02 , RMSE = 4	-0.0012 ± 0.0002 , RMSE = 0.03	–	–
AABW	0.24 ± 0.02 , RMSE = 2	** 0.07 ± 0.01 , RMSE = 2	0.15 ± 0.05 , RMSE = 11	–	–	–

are considered along with changes in the aragonite saturation depth and C_{ANT} storage.

4.1 Changes in DIC, C_{ANT} and pH_T (1995–2011).

4.1.1 Upper ocean layers (STCW, AASW and AASW_{upw})

In the STCW layer, DIC increased between 1995 and 2011 (Fig. 2a, b) at a rate of $0.86 \pm 0.07 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$ ($\frac{\partial \text{DIC}}{\partial t}$, Table 4), leading to a decrease in pH_T of $-0.0027 \pm 0.0001 \text{ yr}^{-1}$ ($\frac{\partial \text{pH}_T}{\partial t}$, Table 4, Fig. 2e, f). The trend in pH_T is similar to the one found by Lauvset et al. (2015) between 1991 and 2011 for the IO-STPS (Indian Ocean subtropical permanently stratified) biome ($-0.0027 \pm 0.0005 \text{ yr}^{-1}$). We found a decrease in DIC^π ($\frac{\partial \text{DIC}^\pi}{\partial t} = -0.34 \pm 0.06 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$, Table 5) in the STCW layer due to a negative trend of $\text{DIC}_{\text{SAT}}^\pi$ resulting

from a decrease in solubility that resulted from a temperature increase (calculated from the section data) in the STCW layer of $0.0335 \pm 0.0130 ^\circ\text{C yr}^{-1}$ (not shown). The increase in temperature agrees with the warming trend observed south of Tasmania of 0.2 to 0.3 $^\circ\text{C decade}^{-1}$, obtained from satellite data (Armour and Bitz, 2015) and from combined data and models (0.5 $^\circ\text{C}$ every 30 years, Aoki et al., 2015). For $\theta = 16 ^\circ\text{C}$ and $S = 35.1$ (definition of $\text{SWT}_{\text{STW16}}$ in the OMP analysis, Table S1), a change in temperature of $0.03 ^\circ\text{C yr}^{-1}$ would lead to a decrease in $\text{DIC}_{\text{SAT}}^\pi$ of $-0.27 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$, which is similar to the value obtained for $\frac{\partial \text{DIC}^\pi}{\partial t}$ in the STCW layer (Table 5). The difference between these trends is related to the mixing of the different SWT fractions within the STCW layer, established from the OMP analysis (see Sect. 3 and Sect. S2). When the solubility change is incorporated into $\frac{\partial \text{C}_{\text{ANT-BC}}}{\partial t}$, i.e. $\frac{\partial \text{C}_{\text{ANT-BC}}}{\partial t} + \frac{\partial \text{DIC}^\pi}{\partial t}$ (Table 5, Fig. 2c, d), we obtain a value of $0.71 \pm 0.08 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$.

Table 5. Trends of anthropogenic carbon from estimates of the back-calculation method and of the terms DIC^{BIO} and DIC^{π} (see Sect. 3.2 of the text). RMSE = root mean square error. Bottom: schematic of the estimation of the upper and lower limits of $\frac{\partial C_{\text{ANT}}}{\partial t}$ (see Sect. 3.3).

Layer	$\frac{\partial C_{\text{ANT_BC}}}{\partial t}$ ($\mu\text{mol kg}^{-1} \text{ yr}^{-1}$)	$\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$ ($\mu\text{mol kg}^{-1} \text{ yr}^{-1}$)	$\frac{\partial \text{DIC}^{\pi}}{\partial t}$ ($\mu\text{mol kg}^{-1} \text{ yr}^{-1}$)
	1995–2011	1995–2011	1995–2011
STCW	1.05 ± 0.05 , RMSE = 5	0.22 ± 0.08 , RMSE = 8	-0.34 ± 0.06 , RMSE = 6
AASW	0.71 ± 0.06 , RMSE = 9	0.50 ± 0.16 , RMSE = 23	-0.36 ± 0.13 , RMSE = 17
AASW _{upw}	–	0.41 ± 0.16 , RMSE = 28	–

Upper limit of $\frac{\partial C_{\text{ANT}}}{\partial t}$
 $(\frac{\partial C_{\text{ANT_BC}}}{\partial t} + \frac{\partial \text{DIC}^{\pi}}{\partial t} + \frac{\partial \text{DIC}^{\text{BIO}}}{\partial t})$

Change due to $\frac{\partial \text{DIC}^{\pi}}{\partial t}$

Change due to $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$

Lower limit of $\frac{\partial C_{\text{ANT}}}{\partial t}$
 $(\frac{\partial C_{\text{ANT_BC}}}{\partial t} + \frac{\partial \text{DIC}^{\pi}}{\partial t})$

Time

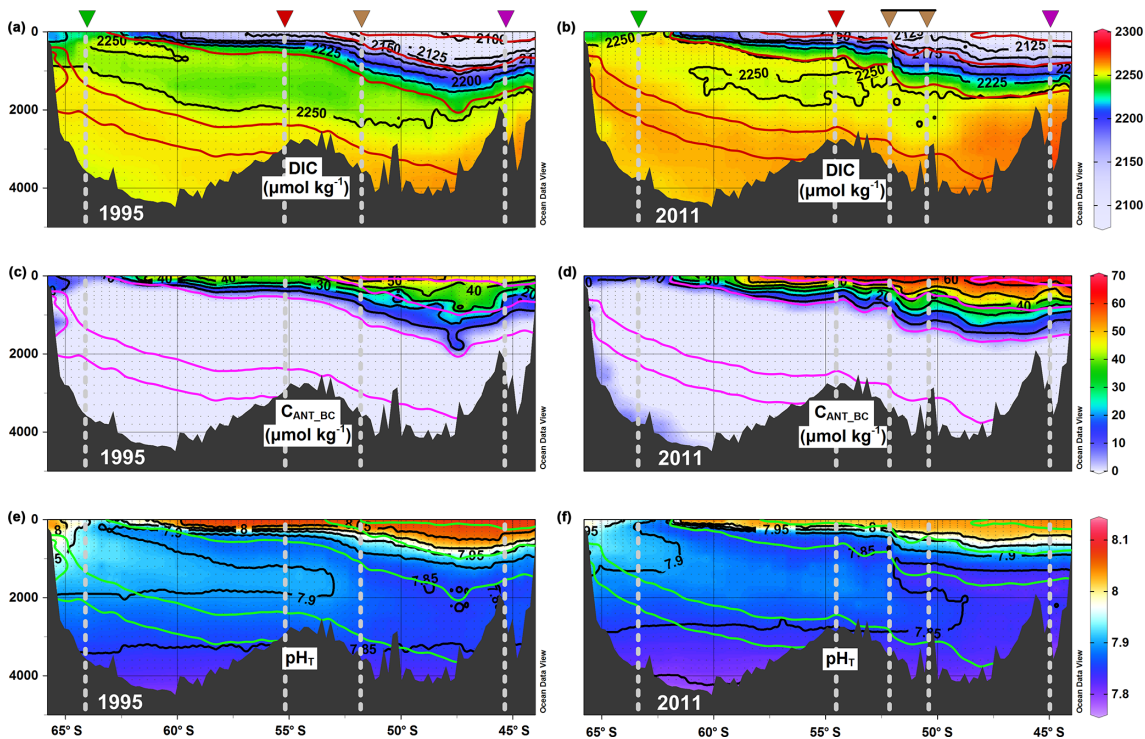


Figure 2. Distribution of DIC (a, b), $C_{\text{ANT_BC}}$ (c, d) and pH_T (e, f) in the SR03 section south of Tasmania for the years 1995 (a, c, e) and 2011 (b, d, f). Red lines in plots (a) and (b), pink lines in plots (c) and (d), and green lines in plots (e) and (f) indicate the neutral surfaces that define the different water masses (γ^n). The position of the fronts (coloured triangles and grey dotted lines) in each of the cruises is also shown.

There is an increase in DIC^{BIO} in the STCW layer ($\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$, Table 5) that also affects the estimates of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$. The increase in DIC^{BIO} is due to an increase in AOU (no changes were found in TA, Eq. 2 in Sect. 3.2) due to a decrease in O_2 in the layer. We cannot separate the effects of circulation and biology on the AOU change, and $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$ in Table 4 should be considered a range. If the changes in AOU are only due to the variability in the remineralisation rates, the calculated lower limit of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$ is $0.71 \pm 0.08 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ (Table 4, $\frac{\partial \text{C}_{\text{ANT}}}{\partial t} = \frac{\partial \text{C}_{\text{ANT_BC}}}{\partial t} + \frac{\partial \text{DIC}^{\pi}}{\partial t}$ in Table 5). If the changes in AOU are due to changes in circulation, the upper limit value of $0.93 \pm 0.11 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ($\frac{\partial \text{C}_{\text{ANT}}}{\partial t} = \frac{\partial \text{C}_{\text{ANT_BC}}}{\partial t} + \frac{\partial \text{DIC}^{\pi}}{\partial t} + \frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$) will explain the increase in DIC in the STCW layer ($\frac{\partial \text{DIC}}{\partial t} \approx \frac{\partial \text{C}_{\text{ANT}}}{\partial t}$, Table 4). The increase in C_{ANT} found in this layer is comparable to the range of increases ($0.8\text{--}1.3 \mu\text{mol kg}^{-1} \text{yr}^{-1}$) found by Carter et al. (2017) in the Pacific Ocean (P16 WOCE, CLIVAR and GOSHIP lines) for the past 2 decades (1990s–2000s and 2000s–2010s).

Changes in the AASW layer are summarised in Table 4. DIC increased at a similar rate of $0.85 \pm 0.14 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ to the STCW layer and the trend is similar to the values found by Williams et al. (2015) for the AASW layer in the Pacific sector of the Southern Ocean ($12\text{--}18 \mu\text{mol kg}^{-1}$ for the period 1992–2011 and $3\text{--}5 \mu\text{mol kg}^{-1}$ for the period 2005–2011). The increase in DIC in the AASW layer results in a pH_T decrease of $-0.0035 \pm 0.0002 \text{yr}^{-1}$, close to Williams et al. (2015) estimates for surface waters ($\sim -0.0023 \pm 0.0009 \text{yr}^{-1}$) and Lauvset et al. (2015) estimates of $-0.0021 \pm 0.0002 \text{yr}^{-1}$ for the Southern Ocean seasonally stratified, SO-SPSS, biome. The AASW layer for our sections warmed at a similar rate ($0.0369 \pm 0.0109 ^\circ\text{C yr}^{-1}$) to the STCW layer, reducing the solubility and influencing $\frac{\partial \text{DIC}^{\pi}}{\partial t}$ (Table 5) due to changes in $\text{DIC}_{\text{SAT}}^{\pi}$. The DIC^{BIO} also increased with time ($0.50 \pm 0.16 \mu\text{mol kg}^{-1} \text{yr}^{-1}$, Table 5) due to an increase in AOU. Following the same reasoning as for the STCW layer and considering the trend in C_{ANT} of $0.70 \pm 0.06 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ obtained by the back-calculation method ($\frac{\partial \text{C}_{\text{ANT_BC}}}{\partial t}$; Table 5), the best estimation of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$ in the AASW layer is a range of 0.35 ± 0.14 to $0.85 \pm 0.22 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ (Table 4). Our values are within the range of values found by Williams et al. (2015) for the AASW in the Pacific sector between 2005 and 2011, and the upper limit is similar to a C_{ANT} increase of $0.73\text{--}0.86 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ for waters South of Tasmania for the period 1968–1996 found by McNeil et al. (2001).

DIC in the AASW_{upw} layer increased at a rate of $0.61 \pm 0.10 \mu\text{mol kg}^{-1} \text{yr}^{-1}$, and the pH_T decreased by $-0.0015 \pm 0.0004 \text{yr}^{-1}$ (Table 4). We were not able to detect a statistically significant trend in DIC^{π} (i.e. solubility) or C_{ANT} from the estimates of the back-calculation

method ($\frac{\partial \text{C}_{\text{ANT_BC}}}{\partial t}$ Table 5). However, we found an increase in DIC^{BIO} of $0.42 \pm 0.28 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ (Table 5) that is due to an increase in AOU. Considering the different drivers of the AOU increase (e.g. biology, circulation), the optimal estimation of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$ for this layer is a value between 0 and $0.42 \pm 0.28 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ (Table 4).

4.1.2 Mode waters and intermediate layers (SAMW and AAIW)

The increase in DIC in the SAMW layer ($1.10 \pm 0.14 \mu\text{mol kg}^{-1} \text{yr}^{-1}$) for the period 1995–2011 is higher than that of upper ocean layers, and pH_T decreases over the same period by $-0.0031 \pm 0.0003 \text{yr}^{-1}$ (Table 4). The DIC increase is explained almost entirely by $\frac{\partial \text{C}_{\text{ANT_BC}}}{\partial t}$ of $0.92 \pm 0.09 \mu\text{mol kg}^{-1} \text{yr}^{-1}$. No significant trend was found in DIC^{BIO} or DIC^{π} (i.e. $\frac{\partial \text{C}_{\text{ANT}}}{\partial t} = \frac{\partial \text{C}_{\text{ANT_BC}}}{\partial t}$). In the AAIW layer the DIC trend of $0.40 \pm 0.15 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ results in a pH_T decrease of $-0.0017 \pm 0.0002 \text{yr}^{-1}$ and is also explained by the increase in C_{ANT} ($0.42 \pm 0.06 \mu\text{mol kg}^{-1} \text{yr}^{-1}$, $\frac{\partial \text{C}_{\text{ANT}}}{\partial t} = \frac{\partial \text{C}_{\text{ANT_BC}}}{\partial t}$, Tables 4 and 5). As with SAMW, no changes in solubility ($\frac{\partial \text{DIC}^{\pi}}{\partial t}$) or biology/circulation processes ($\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$) were detected in the AAIW layer. The values found in the SAMW and AAIW layers are very similar to the mean decadal changes found by Murata et al. (2007) between the 1990s and the 2000s in the subtropical Pacific Ocean ($\sim 1 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ for the SAMW layer and $0.4 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ for the AAIW). Waters et al. (2011) used data from the P18 line along $\sim 110^\circ \text{W}$ and estimated an increase in C_{ANT} of $0.89 \pm 0.4 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ for the SAMW and $0.64 \pm 0.2 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ in AAIW for the period 1994–2008, which are also comparable to our results.

4.1.3 Deep-bottom layers (UCDW, LCDW and AABW)

The UCDW layer shows an increase in DIC of $0.29 \pm 0.02 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ between 1995 and 2011 and a change in pH_T of $-0.0013 \pm 0.0001 \text{yr}^{-1}$, which is similar to the change in DIC ($0.20 \pm 0.02 \mu\text{mol kg}^{-1} \text{yr}^{-1}$) and pH_T ($-0.0012 \pm 0.0002 \text{yr}^{-1}$) for LCDW. No statistically significant changes in time were detected for $\text{C}_{\text{ANT_BC}}$ or in the DIC^{BIO} and DIC^{π} terms for any of these two layers.

The AABW layer also shows an increase in DIC ($0.24 \pm 0.02 \mu\text{mol kg}^{-1} \text{yr}^{-1}$) during the period 1995–2011 with an associated decrease in pH_T of $-0.0013 \pm 0.0002 \text{yr}^{-1}$ (Table 4). The increase in C_{ANT} ($\frac{\partial \text{C}_{\text{ANT_BC}}}{\partial t}$) of $0.07 \pm 0.01 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ is low and this trend indicates an increase in C_{ANT} of $\sim 1 \mu\text{mol kg}^{-1}$, which is less than the accuracy of the back-calculation method ($\pm 6 \mu\text{mol kg}^{-1}$).

Table 6. Approximated rates of C_{ANT} storage based on the trends from Table 4.

Layers	Thickness (m)	C_{ANT} storage rates ($\text{mol m}^{-2} \text{yr}^{-1}$)
STCW	230	$[0.17 \pm 0.06 - 0.22 \pm 0.08]$
AASW	280	$[0.10 \pm 0.04 - 0.24 \pm 0.07]$
AASW _{upw}	275	$[0 - 0.11 \pm 0.04]$
SAMW	450	0.43 ± 0.14
AAIW	900	0.39 ± 0.14
SR03 section	–	0.30 ± 0.24

4.2 Changes in the aragonite saturation ($\frac{\partial \Omega_{\text{Ar}}}{\partial t}$) and C_{ANT} storage

There are statistically significant decreases in Ω_{Ar} in the STCW, AASW and SAMW layers ($\sim -0.010 \pm 0.001 \text{ yr}^{-1}$, Table 4) similar to the trends observed at open-ocean time series sites in recent decades (Bates et al., 2014). The decrease in Ω_{Ar} found for the AASW layer ($-0.61 \pm 0.19 \% \text{ yr}^{-1}$, Table 4) is also similar to the values obtained by Williams et al. (2015) for the Pacific sector of the Southern Ocean ($-0.47 \pm 0.10 \% \text{ yr}^{-1}$ for the period 1992–2011 and $-0.50 \pm 0.20 \% \text{ yr}^{-1}$ for the period 2005–2011). Accompanying the decrease in Ω_{Ar} with time along SR03 is the shoaling of the aragonite saturation depth (ASD, $\Omega_{\text{Ar}} = 1$, Fig. 3) at a mean rate of $-13 \pm 3 \text{ m yr}^{-1}$. The shoaling of the ASD is not uniform over the section. North of the PF, the ASD shoals at a rate of $-6 \pm 4 \text{ m yr}^{-1}$ while the rate is 3.5 times greater south of the PF ($-21 \pm 4 \text{ m yr}^{-1}$). North of the PF the shoaling mostly affects the AAIW layer (Fig. 3a). South of the PF, from $\sim 62^\circ \text{ S}$, the movement of the ASD follows the upwelling path of the UCDW layer (Fig. 3) with a shoaling of $\sim 340 \text{ m}$ over the 16-year period.

The storage rate of C_{ANT} (Table 6) for the surface and intermediate water mass layers is obtained from $\frac{\partial C_{\text{ANT}}}{\partial t}$ (Table 4), with the most storage in SAMW and AAIW due to both their greater thickness and $\frac{\partial C_{\text{ANT}}}{\partial t}$ values. The rate of increase of the C_{ANT} storage in the whole longitude band of the SR03 section is $0.30 \pm 0.24 \text{ mol m}^{-2} \text{ yr}^{-1}$, calculated by computing the mean of the storage rates of the layers weighted by the mean volume occupied by each of the layers for the period 1995–2011 (Table 6)

5 Discussion

Our results are indicative of a scenario of increased transport of deep waters into the section and enhanced upwelling at high latitudes for the period between 1995 and 2011 linked to strong westerly winds. Several studies have reported a trend in the Southern Annular Mode (SAM) toward its positive phase from the 1960s until the 2000s (Thompson and Solomon, 2002; Marshall, 2002, 2003; Lenton and Matear,

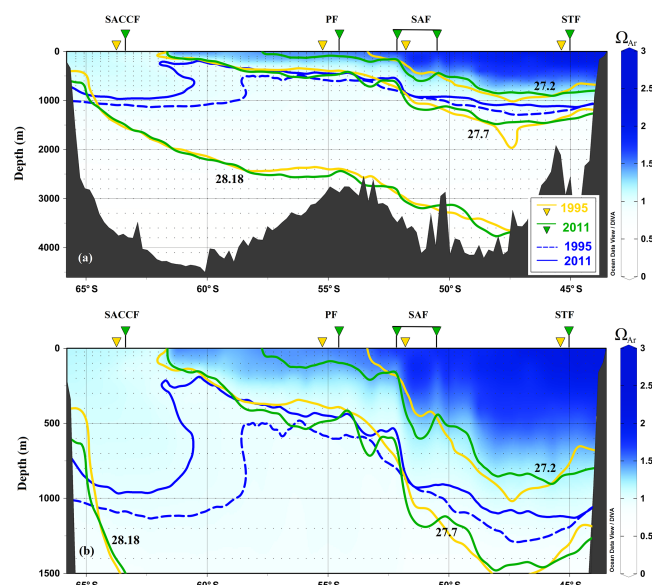


Figure 3. Aragonite saturation depth (ASD, $\Omega_{\text{Ar}} = 1$) for the 1995 and 2011 cruises. (a) Distribution of ASD (blue dotted and solid lines) and γ^n (green and yellow lines) in latitude in the SR03 section. (b) Section zoomed for the first 1500 m of the water column. The position of the fronts (triangles) in 1995 (yellow) and 2011 (green) is also shown. The blue palette in the background indicates the distribution of Ω_{Ar} for the 2011 cruise.

2007; Sallée et al., 2008). According to these studies, the positive phase of the SAM is correlated with an intensification and southward movement of the subpolar westerly winds that ultimately lead to the enhancement of northward Ekman transport, meridional overturning and upwelling south of the ACC. Also, surface warming and more intense and frequent pulses in the extension of the EAC on long timescales have been related to a poleward movement of the westerly winds (Rintoul and Sokolov, 2001; Ridgway, 2007; Hill et al., 2011). From the 2000s on, the SAM index no longer presents a positive trend but, despite exhibiting considerable inter-annual variability (Fig. S2), remains in its positive phase, favouring strong winds over the region.

In the northern part of the SR03 section, the area occupied by the STCW has high variability due to the encounter between the EAC and the ZC in the north of the section (Ridgway et al., 2007; Herraiz-Borreguero and Rintoul, 2011; Sloyan et al., 2016). The warming of the STCW layer found in this study ($0.0335 \pm 0.0130 ^\circ \text{C yr}^{-1}$) could be linked to variability in the extension of subtropical waters but it could also be related to atmospheric warming. Aoki et al. (2015) related the 30-year warming found north of the SAF in the South Pacific and Indian oceans to the intensification of the subtropical gyres, which promote the arrival of warmer waters. In the AASW layer that extends approximately between the SAF and the PF we found a similar warming ($0.0369 \pm 0.0109 ^\circ \text{C yr}^{-1}$) to that of the STCW.

This could indicate that the increase in temperature found in the upper layers of the section could be most likely due to ocean heat uptake and atmospheric warming.

Due to the surface warming, the increase in DIC found in the STCW layer (Table 4) is lower than expected from the increase in atmospheric CO₂ ($\sim 1 \pm 0.12 \mu\text{mol kg}^{-1} \text{yr}^{-1}$). Nevertheless, at least 83 % of the increase in DIC in the STCW layer is explained by the increase in C_{ANT} (Table 4). As for the AASW layer, our results indicate that temperature does affect the estimate of $\frac{\partial \text{DIC}}{\partial t}$, but the effect of the increase in DIC^{BIO} (due to an increase in AOU) outweigh that of solubility (Table 5).

The seasonal to inter-annual variability of the AASW layer is also influenced by the variability of the positions of the SAF and PF (Fig. 1, Table 3), which is highly conditioned by the flow of the ACC over the South-East Indian Ridge (Fig. 1). A close relationship between phytoplankton blooms and regions where the ACC fronts interact with large topographic features has been noticed (Moore et al., 1999; Moore and Abbott, 2000). A variability in the remineralisation rates due to phytoplankton bloom variability could explain the changes in DIC^{BIO} observed in the AASW layer. Nevertheless, no changes in nutrients (nitrates or phosphates) are measurable in this layer that could indicate intense biological activity.

Furthermore, the AASW layer is also affected by the upwelling of deep waters south of the PF, and an intensification of the upwelling could increase the content of low-O₂ DIC-rich waters in the AASW layer, leading to an increase in AOU. The increase in DIC^{BIO} found in the AASW_{upw} layer (Table 4), south of the PF, is similar to the increase obtained for the AASW layer, which indicates the likelihood that the upwelling of deep waters results in the increase in AOU. The increase in DIC^{BIO} in the AASW_{upw} layer coincides with an increase in salinity of $0.0029 \pm 0.0001 \text{yr}^{-1}$ (not shown), which is consistent with increased transport of saltier waters from the deep ocean to subsurface layers. Besides, we also found an increase in dissolved silicate of $0.36 \pm 0.06 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ($\frac{\partial \text{SiO}_4}{\partial t}$ Table 4) that could be related to the upwelling enhancement as well (Tréguer, 2014).

The influence of the upwelling on the DIC budgets (as non-anthropogenic DIC) is clearer in the AASW_{upw} layer than in the AASW layer. For the AASW layer, the lower limit of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$ (i.e. the change in DIC^{BIO} is assumed to be due to biological processes, Table 4) indicates that at least 41 % of the increase in DIC in the layer is explained by the increase in C_{ANT} while the lower limit of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$ is zero for AASW_{upw} (Table 4), meaning that the effect of the upwelling over AASW is lower than over AASW_{upw}. Matear and Lenton (2008), using carbon models, concluded that the uptake of CO₂ by the waters north of the PF is more influenced by the wind variability than by other processes such as the upwelling. An intensification of the winds (due to a posi-

tive phase in SAM) could contribute to the increase in C_{ANT} found in the AASW layer. Considering the upper limits of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$ in both layers (i.e. the change in DIC^{BIO} is assumed to be due to circulation processes), the increase in C_{ANT} in the AASW_{upw} layer represents no more than 69 % of the increase in DIC (upper limit of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$, Table 4), while the upper limit of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$ for the AASW equals the increase in DIC. Thus, in the AASW_{upw} layer, at least ~ 30 % of the increase in DIC ($\sim 0.18 \mu\text{mol kg}^{-1} \text{yr}^{-1}$) is still not explained and is most probably related to the upwelling of DIC-rich waters. The increase in non-anthropogenic DIC could be even higher, since we assume that the change in DIC^{BIO} due to circulation does not affect DIC (see Sect. 3.3).

In terms of the change in oxygen, Helm et al. (2011) found an average decrease in the concentration of O₂ between 100 and 1000 m from 1970 to 1992 of $\sim -0.23 \mu\text{mol L}^{-1}$ for the Southern Ocean (27 % of the estimated global average change, $-0.93 \pm 0.23 \mu\text{mol L}^{-1}$). Considering the volume of the first 1000 m of the water column of the Southern Ocean to be $19\,400 \times 10^9 \text{ l}$ (obtained using ETOPO1, Amante and Eakins, 2009) and the volume of the first 1000 m of the SR03 section to be $2700 \times 10^9 \text{ l}$, the decrease in O₂ found by Helm et al. (2011), if constant in time, would correspond to a decrease of $\sim -1.7 \mu\text{mol L}^{-1} \text{yr}^{-1}$. We only found changes in oxygen within the surface water mass layers (STCW, AASW and AASW_{upw}) that approximately fill the first 300 m of the water column of the SR03. Then, the decrease of $\sim -1.7 \mu\text{mol L}^{-1}$ would correspond to an average change in O₂ of $\sim -0.32 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ for surface waters of the SR03. This means that values of $\sim 0.20 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ due to circulation processes can be expected in $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$ for surface waters, which is comparable to the average of our findings (Table 5), $0.32 \pm 0.24 \mu\text{mol kg}^{-1} \text{yr}^{-1}$, and could indicate that the change in O₂ is related to circulation processes.

The variability of the SAMW and AAIW layers south of Tasmania has been related to variability in the northward Ekman transport that drives the northward movement of AASW (Rintoul and England, 2002; Sallée et al., 2006, 2012). A scenario of intensification of the upwelling near the Antarctic Divergence would lead to an increase in the northward Ekman transport, conditioning the properties of these water mass layers, particularly for SAMW, which is mostly formed north of the SAF. There is a significant freshening of the SAMW layer ($-0.0026 \pm 0.0001 \text{psu yr}^{-1}$, not shown) between 1995 and 2011 that could be related to higher inputs of AASW into the SAMW layer and is consistent with the increase in Ekman transport. Besides, an intensification of the winds due to the positive trend of the SAM favours the ventilation and thus the increase in C_{ANT} uptake by both water mass layers (Matear and Lenton, 2008). Our results indicate that the change in DIC in the SAMW and AAIW layers is driven mostly by the uptake of atmospheric CO₂ ($\frac{\partial \text{DIC}}{\partial t} \approx \frac{\partial \text{C}_{\text{ANT}}}{\partial t}$, Table 4). The increase in C_{ANT} in the SAMW layer is higher than that found for the upper ocean

Table 7. Comparison between $\frac{\partial C_{\text{ANT}}}{\partial t}$ from the present study and the values of ΔC_{ANT} from the two-regression and eMLR methods. RMSE = root mean square error. ** The value of $\frac{\partial C_{\text{ANT}}}{\partial t}$ in the AABW layer is considered negligible because it falls below the accuracy of the back-calculation method.

Layer	$\frac{\partial C_{\text{ANT}}}{\partial t}$ ($\mu\text{mol kg}^{-1} \text{ yr}^{-1}$)	ΔC_{ANT} (two-regression) ($\mu\text{mol kg}^{-1} \text{ yr}^{-1}$)	ΔC_{ANT} (eMLR) ($\mu\text{mol kg}^{-1} \text{ yr}^{-1}$)
	1995–2011	1995–2011	1995–2011
STCW	$[0.71 \pm 0.08 - 0.93 \pm 0.08]$	0.72 ± 0.04 , RMSE = 5	1.21 ± 0.09 , RMSE = 3
AASW	$[0.35 \pm 0.14 - 0.85 \pm 0.21]$	0.61 ± 0.04 , RMSE = 6	0.67 ± 0.10 , RMSE = 10
AASW _{upw}	$[0 - 0.41 \pm 0.16]$	0.20 ± 0.03 , RMSE = 3	0.30 ± 0.04 , RMSE = 3
SAMW	0.92 ± 0.09 , RMSE = 10	0.94 ± 0.05 , RMSE = 5	0.96 ± 0.09 , RMSE = 10
AAIW	0.42 ± 0.06 , RMSE = 10	0.28 ± 0.02 , RMSE = 4	0.43 ± 0.02 , RMSE = 7
UCDW	–	0.09 ± 0.01 , RMSE = 2	0.15 ± 0.09 , RMSE = 3
LCDW	–	0.08 ± 0.01 , RMSE = 2	0.28 ± 0.15 , RMSE = 3
AABW	** 0.07 ± 0.01 , RMSE = 2	0.09 ± 0.01 , RMSE = 1	0.18 ± 0.08 , RMSE = 2

layers and closer to the expected values from the increase in atmospheric CO_2 ($\sim 1 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$). The smaller increase in C_{ANT} in the AAIW layer compared to the SAMW layer (Table 4) agrees with lower ventilation of the AAIW layer south of Tasmania (see Sect. 2) due to the fact that this layer carries recently ventilated waters mixed with older waters ventilated far out the SR03 section. The lack of measurable long-term changes in DIC^{BIO} and DIC^{π} in both AAIW and SAMW layers indicate that circulation and biological processes do not have a large effect on $\frac{\partial \text{DIC}}{\partial t}$.

Deep to bottom layers of the section show significant trends for DIC that are not explained by the increase in C_{ANT} . These trends are most likely due to the advection of old and DIC-rich waters. Concretely for deep waters (UCDW and LCDW), the trends could result from an intensification of upwelling at high latitudes being offset by enhanced transport of old and CO_2 -rich waters to replace the upwelled waters, since the increase in DIC follows the upwelling path of the UCDW and LCDW layers (Fig. 4). We separated the UCDW layer into two latitudinal sectors: north and south of the SAF (Fig. 4). The increase in DIC in the UCDW layer north of the SAF is $0.44 \pm 0.04 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$ while that south of the SAF is smaller at $0.26 \pm 0.04 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$ (not shown), consistent with a greater supply of waters from the north at depth. A decrease in O_2 in the UCDW to the north of the SAF occurs mostly in the upper to middle parts of the UCDW layer (Fig. 4), and this is not observed south of the SAF. The decrease in O_2 is also in agreement with the arrival of waters from Indian–Pacific origin since these waters provide the characteristic oxygen minimum zone that defines UCDW (Callahan, 1972; Talley 2013). Another feature that agrees with the hypothesis of upwelling intensification is the shoaling of the ASD following the path of upwelling of the UCDW layer. This feature was also described by Bostock et al. (2013) in an oceanic climatology of Ω_{Ar} and could be due to the naturally lower buffer capacity of the UCDW layer (low value of $\text{TA} / \text{DIC} \approx 1.043$) with respect to upper layers

($\text{TA} / \text{DIC} \approx 1.06$ in the AASW layer). However, the greatest shoaling of the ASD in the UCDW layers compared to the AAIW layer (Table 6) is consistent with the upwelling of UCDW, as both water masses have similar TA / DIC ratios ($\text{TA} / \text{DIC} \approx 1.043$ for the UCDW layer and $\text{TA} / \text{DIC} \approx 1.042$ for the AAIW layer). Furthermore, the increase in SiO_4 found in deep-bottom layers (Table 4) could also indicate the arrival of old waters to the section that are progressively enriched in SiO_4 (e.g. Callahan, 1972).

Statistically significant decreases in pH_T with time were observed in all water mass layers (Table 4), with the greatest change in surface water masses coinciding with the greatest DIC changes. The decrease in pH_T in the STCW and SAMW layers is related to the increase in the uptake of C_{ANT} , while for the AASW and AASW_{upw} layers the pH_T change appears to be linked to the upwelling of DIC-rich waters at high latitudes. At deep layers, the tongue of water of $\text{pH}_T = 7.9$ off the shelf is reduced in 2011 compared to 1995 (Fig. 2e, f), which is consistent with the advection of DIC-rich waters in the section due to the enhanced upwelling. The different rates of pH_T change in the water masses is in part related to the buffering capacity of the waters. The AASW layer has lower temperature than the STCW layer ($\sim 2.3^\circ\text{C}$ for the AASW layer compared to $\sim 11.0^\circ\text{C}$ for the STCW layer, mean values for the period 1995–2011) and lower buffer capacity than the STCW ($\text{TA} / \text{DIC} \approx 1.058$ for the AASW layer versus $\text{TA} / \text{DIC} \approx 1.095$ for STCW). For similar increases in DIC (Table 4) the decrease in pH_T in the AASW is expected to be higher than in the STCW layer.

In terms of carbon, previous studies concluded that the intensification of the upwelling (as a consequence of the SAM variability) caused a reduction in the uptake of CO_2 by the Southern Ocean between the 1980s and 2000s due to the outgassing of CO_2 near the Antarctic Divergence (Le Quéré et al., 2007; Lovenduski et al., 2008). Landschützer et al. (2015) showed that the efficiency of the Southern Ocean CO_2 sink declined through the 1990s, and the trend reversed

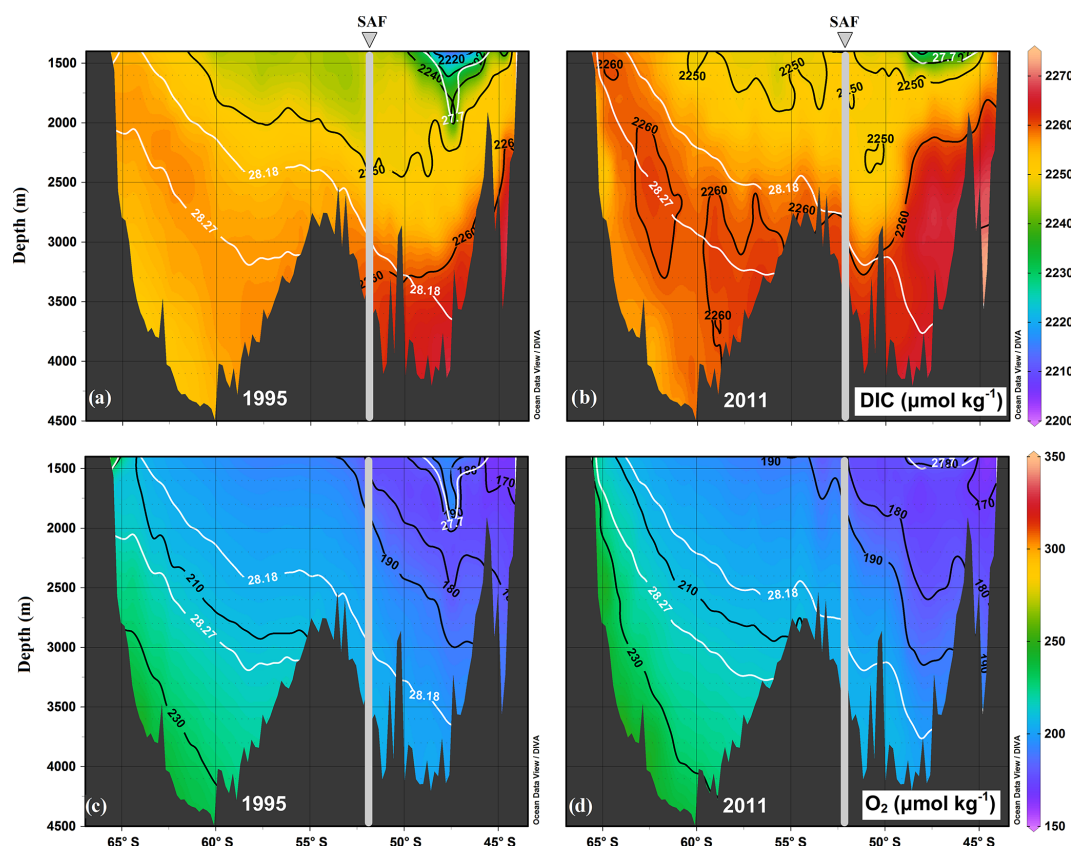


Figure 4. Distribution of (a, b) dissolved inorganic carbon (DIC) and (c, d) dissolved oxygen (O_2) in the 1995 and 2011 cruises for deep-bottom layers of the section. The location of the Sub-Antarctic Front (SAF) is also shown as well as the neutral surfaces (γ^n , white lines) limiting the UCDW, LCDW and AABW layers.

from about 2002, although the reversal in the sink efficiency was not zonally uniform. The results from Landschützer et al. (2015) are consistent with a carbon sink influenced by the upwelling of DIC-rich waters at high latitudes, and superimposed on this is the near-surface response to atmospheric forcing that modifies the sink efficiency and could mask longer-term trends in the upwelling of DIC-rich waters at high latitudes. A comparison of our results with those of Landschützer et al. (2015) is problematic as their data are restricted to surface waters and our analysis is on long-term trends in water mass properties below 50 m depth. Both data sets do show continued uptake of CO_2 throughout the period of study and indicate the importance of the circulation in influencing the regional carbon sink, which has also been established by recent model results (DeVries et al., 2007).

Our results also agree with the conclusions from different model simulations done by Matear and Lenton (2008), who established that intense wind regimes (associated with a positive phase in the SAM) favour the uptake of C_{ANT} and ventilation of the SAMW and AAIW layers. These authors highlighted the complex response of the uptake of CO_2 by the Southern Ocean due to the diverse forcing acting on upper layers, which can be also seen in our results (e.g. differences

in the biogeochemical changes in the AASW and AASW_{upw} layers). Matear and Lenton (2008) also noticed the complex relationship between the upwelling and subduction areas of the Southern Ocean, with the same drivers acting in opposite directions for the changes in non-anthropogenic DIC with respect to the changes in C_{ANT} uptake.

6 Sensitivity of the results to underlying assumptions

This section considers the sensitivity of assumptions used to calculate temporal changes in C_{ANT} , including errors associated with the assumption of steady state in the oceanic circulation and remineralisation processes, and the sensitivity to stoichiometric ratios for the biological processes.

6.1 Comparison of C_{ANT} changes using other methods

We compared the changes in C_{ANT} obtained in our study with the results from two regression-based methodologies (Table 7): the extended multiple linear regression (eMLR) method (Friis et al., 2005) and the two-regression method (Thacker, 2012). These methods use repeated hydrodynamic sections to quantify the temporal change in C_{ANT} .

The eMLR method (Friis et al., 2005) estimates the change in C_{ANT} between two repeats of a hydrodynamic section by establishing MLRs for each section and relating the observed DIC for each observation to a set of other measured oceanic variables:

$$\text{DIC}_{(t)} = a_{0(t)} + a_{1(t)}P_{1(t)} + \dots + a_{n(t)}P_{n(t)}, \quad (6)$$

where $a_{x(t)}$ are the coefficients of the fit between DIC and the n observed variables (P_1, \dots, P_n) chosen for the fit, all measured at the time (t) of the survey.

Taking the difference between DIC at two times, t_1 and t_2 , gives an equation for the change in C_{ANT} over the time period between the two hydrographic surveys (ΔC_{ANT}):

$$\Delta C_{\text{ANT}} = a_{0(t_2)} - a_{0(t_1)} + (a_{1(t_2)} - a_{0(t_1)})P_{1(t_2)} + \dots + (a_{n(t_2)} - a_{0(t_1)})P_{n(t_2)} \quad (7)$$

The two-regression method was introduced by Thacker (2012) as an improvement in regression-based methods. The region of study is first divided into sub-regions since the empirical relationships between DIC and other environmental variables vary spatially (Thacker, 2012). MLRs are investigated between DIC and other measured variables (predictors) using a stepwise technique. The procedure is applied for each sub-region using all data from the repeated surveys within the period to be investigated, resulting in an optimal MLR for each sub-region (similar to Eq. 6). A linear regression with time is established for the residuals (observed DIC – predicted DIC) of the regional fits, which directly gives ΔC_{ANT} averaged over the space–time in each sub-region. The purpose of the first MLR is to remove the natural variability of DIC, leaving the anthropogenic signal and noise (random variability) in the residuals, and the second MLR is used to separate the anthropogenic signal from the noise.

We applied both methodologies within the different water mass layers, which were separated by γ^n and used as sub-regions. The predictor variables of θ , S , σ_0 , nitrate (NO_3), SiO_4 and AOU were used for the MLR procedures. The three methodologies estimate similar rates of increase in C_{ANT} for most water mass layers (Table 7). In the STCW layer, the value of ΔC_{ANT} (eMLR) is higher than our maximum estimate of $\frac{\partial C_{\text{ANT}}}{\partial t}$ and the value obtained from the two-regression method. The eMLR method is less suitable for the upper layers of the ocean subject to high seasonal to inter-annual variability, such as the STCW layer, resulting in large residuals that bias the regression (Friis et al., 2005). For the AAIW layer, the increase in C_{ANT} estimated by the two-regression method is half the increase that is established by our method and the eMLR method. The lower value of the trend estimated by the two-regression method is due to the fact that the two-regression method uses stepwise MLR. This means that the two-regression method only considers those predictors that give the best fit while the eMLR method is forced to consider all the predictors in the

fit. This is also the cause of the low RMSE obtained with the two-regression method compared to both our trends and those obtained by the eMLR method. For deep to bottom layers, the two-regression method estimates a small increase in C_{ANT} similar to the one found in this study for the AABW layer (Table 7) that can be considered negligible given the resolution of the back-calculation method ($\pm 6 \mu\text{mol kg}^{-1}$). The eMLR method finds increases in C_{ANT} (with relatively high uncertainties) higher than the two-regression method that over the 16-year period also give values of C_{ANT} change lower than the resolution of our back-calculation method (although close to it for the LCDW layer, $\sim 5 \mu\text{mol kg}^{-1}$ for the 16-year period).

6.2 Circulation and biological processes at steady state

The back-calculation method assumes the circulation of the ocean and the biological processes are at steady state. The contribution of non-linear mixing is unknown, and some of the changes in DIC in the water mass layers could be erroneously included in the estimates of C_{ANT} rather than as a non-anthropogenic change in DIC. The non-steady state of the circulation in our analysis is included to some extent through the changes in CDIS^π (Eqs. 3, 4), which is solved by the OMP analysis, which is subjected to the limitations of quantifying the mixing mostly through thermohaline changes in the water masses (Sect. S2).

For biological processes, remineralisation rates are usually considered to be at steady state (Sarmiento et al., 1992). Climate change has been suggested as potentially driving changes in carbon fixation and export that can influence the uptake of CO_2 by the oceans (Falkowski et al., 1998). Pahlow and Riebesell (2000) first suggested that decadal changes in remineralisation rates occurred in the deep waters of the Northern Hemisphere, although this is still a matter of debate (e.g. Li and Peng, 2002; Najjar, 2009).

Metzl et al. (1999) and Shadwick et al. (2015) observed that the uptake of CO_2 over the sub-Antarctic zone (SAZ, between the SAF and the STF) in summer is mostly controlled by biological processes. If a change in remineralisation rates has occurred, i.e. the changes in DIC^{BIO} are due to biological effects, a change in nutrient concentrations of the water masses would be expected. The detection of long-term trends of nutrients in upper layers of the ocean can be masked by short-timescale physical processes such as changes in the mixed layer depth, mesoscale activity and advection (Sambrotto and Mace, 2000; Rintoul and Trull, 2001; Sallée et al., 2010). We did not find a measurable trend in nutrient concentrations for any of the layers in the period 1995–2011, except for an increase in SiO_4 over the Antarctic Divergence (Table 4) that is most likely due to the upwelling of SiO_4 -rich deep waters. We cannot confidently assign the changes in DIC^{BIO} and its impact on $\frac{\partial C_{\text{ANT}}}{\partial t}$ in upper layers (Table 5) to any particular process, and instead we provide a range of values for $\frac{\partial C_{\text{ANT}}}{\partial t}$ (Table 4). For the scenario of intensification

of the Antarctic upwelling, the increase in low- O_2 and DIC-rich waters would increase the content of DIC in subsurface waters, leading (at least for the AASW and AASW_{upw} layers) to values of $\frac{\partial \text{DIC}}{\partial t}$ closer to the lower limit of the range (i.e. $\frac{\partial \text{DIC}}{\partial t} \gg \frac{\partial \text{C}_{\text{ANT}}}{\partial t}$, Table 4).

In deep layers of the section, the increase in DIC is not explained by the long-term change in any of the terms in Eq. (1), which is another implication of considering the circulation at steady state. The differences found in the increase in DIC in the UCDW layer north and south of the SAF (Sect. 4.3, Fig. 4) add consistency to the idea of the advection of older waters to the section. Considering these differences, we can assign a change in DIC of at least $\sim 0.20 \pm 0.02 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ (lower rate of increase in DIC found in deep–bottom layers, Table 4) due to the upwelling intensification. Since the AASW and AASW_{upw} layers are the most affected by the upwelling, we can correct the values of $\frac{\partial \text{C}_{\text{ANT}}}{\partial t}$ in these layers for this effect (trends with ** in Table 4).

6.3 Changes in the rates of export of particulate organic carbon and silicate from surface layers

We assume that the export of particulate organic carbon (POC), from upper ocean layers (STCW, AASW and AASW_{upw}) and remineralisation in the water column, was constant between 1995 and 2011. The high latitudes are considered important in terms of POC export, mostly because these areas are dominated by large phytoplankton, in particular diatoms (Buesseler, 1998; Sambrotto and Mace, 2000), and rapid export of carbon to deep waters from phytoplankton blooms is possible (DiTullio et al., 2000; Lourey and Trull, 2001). The POC exported is remineralised to DIC below the mixed layer (Wassman et al., 1990; Asper and Smith Jr., 1999; Trull et al., 2001a; Fripiat et al., 2015).

Our results show that the increase in DIC in mode and intermediate waters is fully explained by the uptake of atmospheric CO_2 , which could indicate that there was no detectable change in the rate of export of POC over the 1995–2011 period. Estimates of POC export in the SAZ ($\sim 3800 \text{ m}$), the SAF ($\sim 3100 \text{ m}$) and the PFZ ($\sim 1500 \text{ m}$) using moored sediment traps near the section (Trull et al., 2001b) were 0.5, 0.8 and $1.0 \text{ g C m}^{-2} \text{yr}^{-1}$, respectively. If all this POC is fully remineralised in the UCDW layer (with a mean thickness of 2000 m), we obtain a range of $0.02\text{--}0.04 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ for the maximum increase in DIC due to the export of POC. This increase is close to the uncertainty of the total DIC increase estimated for the UCDW layer, which means that in order to generate an increase in DIC similar to that found in the UCDW layer, the rate of POC export should be ~ 10 times higher than the observed rates. This change should certainly be noticeable in $\frac{\partial \text{DIC}^{\text{BIO}}}{\partial t}$ in surface waters but most probably in deep waters as well, which we do not see.

The observed increase in SiO_4 in deep and bottom layers of the ocean is consistent with the transport of SiO_4 -rich older waters to the section. For UCDW, the increase in SiO_4 north of the SAF is higher than at southern latitudes ($0.31 \pm 0.08 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ with respect to $0.19 \pm 0.04 \mu\text{mol kg}^{-1} \text{yr}^{-1}$, not shown). Nelson et al. (1995) observed a lower dissolution rate of diatom-dominated SiO_4 exported in high-latitude regions compared to lower latitudes. Nelson et al. (1995) estimated a mean silica production rate of $0.7\text{--}1.2 \text{ mol Si m}^{-2} \text{yr}^{-1}$ for regions over diatomaceous sediments and concluded that 15–25 % of the silica produced in the upper ocean accumulates in the seabed. Of the silica produced in the mixed layer of the upper ocean layers, at least 50 % is believed to dissolve in the upper 100 m of the water column (e.g. Nelson et al., 1991; DeMaster et al., 1992). For deep waters, the production rates of Nelson et al. (1995) could result in a mean increase in SiO_4 of $0.08\text{--}0.14 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ in the water column ($\sim 3400 \text{ m}$, mean depth). The maximum value of this increase could explain the trends of SiO_4 found for the AABW layer (Table 4), but 32–48 % of the increase in SiO_4 in deep bottom layers is not explained by the remineralisation of exported silica and is most likely the result of the advection of older SiO_4 -rich waters to the section.

6.4 Stoichiometric ratios for biological processes

The back-calculation method and the OMP analysis assume constant stoichiometric ratios for remineralisation. The theoretical Redfield ratios (Redfield, 1934, 1958) are usually considered as a mean for the whole ocean, although they can vary from the theoretical value due to changes in phytoplankton species composition, the food-web structure and nutrient availability (Martiny et al., 2013).

We carried out a sensitivity analysis on the Redfield ratios following Álvarez et al. (2014), to obtain values of stoichiometric ratios for the section. A battery of OMP analyses were done with varying values of R_N between 9 and 10 in increments of 0.2, R_P between 120 and 145 in increments of 5, and R_{Si} between 0 and 8 in increments of 2. For each variation in the stoichiometric ratios, an OMP analysis was made for each section in order to determine best-fit R values and whether there were differences in time for the stoichiometric ratios along the sections. The smallest residuals (differences between the nutrients measured and those estimated by the OMP analysis) were obtained for $R_N = 9$ and $R_P = 125$. The residuals of SiO_4 did not change significantly for any value of R_{Si} and we consider SiO_4 to be a conservative variable. These results indicate $\frac{R_P}{R_N} = 13.8$, in agreement with values obtained for the region $\left(\frac{R_P}{R_N} \in [8\text{--}15], \text{ Lourey and Trull, 2001}\right)$.

7 Conclusions

The results of our analysis south of Tasmania over the 1995–2011 period support a scenario of intensification of upwelling in the vicinity of the Antarctic Divergence due to an increase in the westerly winds at high latitudes, most probably linked to the variability of the SAM. The intensification of the upwelling favours the advection of older waters to deep–bottom layers of the section where we found a net increase in DIC over the 16-year period. The enhanced upwelling causes the eventual entrainment of low- O_2 and DIC-rich waters into upper layers, explaining the trends of decreasing O_2 and increasing non-anthropogenic DIC found in surface waters close to the Antarctic Divergence. This scenario also implies the intensification of the convergence north of the SAF, implying a more efficient ventilation of the SAMW and AAIW layers and thus an efficient uptake of atmospheric CO_2 by these layers. The enhanced upwelling lowers the uptake of C_{ANT} in the AASW layer, but the effect of ventilation more than compensates that of the upwelling, allowing the increase in C_{ANT} in this layer. The atmospheric warming reduces the dissolution of CO_2 in upper layers north of the PF, presenting increases in DIC lower than expected from the atmospheric CO_2 increase.

Our results rely on a limited number of sections that are studied every 3–7 years and can only provide a long-term (decadal) average view of changes in water masses. More surface observations and repeated deep-ocean sections are needed to help resolve inter-annual changes in the Southern Ocean carbon sink and to determine the main drivers and feedback to the carbon–climate system. The effort to maintain hydrographic sections with CO_2 system measurements would also benefit from additional direct measurements of more variables of the carbon system, e.g. pH, which has not been measured in the SR03 section.

Data availability. The section data are available through the Global Ocean Data Analysis Project (<https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2/>; Key et al., 2015; Olsen et al., 2016). The original data for the different cruises were corrected following the QC recommendations in GLODAPv2.

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Competing interests. The authors declare that they have no conflict of interest.

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