Ocean ventilation controls the contrasting anthropogenic CO_2 uptake rates between the western and eastern South Atlantic Ocean basins

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Key Points:

- Between the 1980s and 2010s, the uptake rate of CO_2 in the South Atlantic Ocean was 3.86±0.14 Pg C decade^{-1}.
- The anthropogenic CO_2 absorbed by the basin west of the Mid-Atlantic Ridge is about three times that of the eastern basin.
- The lower uptake in the eastern basin is attributed to lower ventilation rates.

Abstract

The South Atlantic Ocean is an important region for anthropogenic CO_2 (C_{anth}) uptake and storage in the world ocean, yet is less studied. Here, after an extensive sensitivity test and method comparison, we applied an extended multiple linear regression (eMLR) method with six characteristic water masses to estimate C_{anth} change or increase (ΔC_{anth}) between 1980s and 2010s in the South Atlantic Ocean using two meridional transects (A16S and A13.5) and one zonal transect (A10). Over a period of about 25 years, the basin-wide ΔC_{anth} was 3.86±0.14 Pg C decade^{-1}. The two basins flanking the Mid-Atlantic Ridge had different meridional patterns of ΔC_{anth}, yielding an average depth-integrated ΔC_{anth} in the top 2000 m of 0.91±0.25 mol m^{-2} yr^{-1} along A16S on the west and 0.57±0.22 mol m^{-2} yr^{-1} along A13.5 on the east. The west-east basin ΔC_{anth} contrasts were most prominent in the tropical region (0-20°S) in the Surface Water (SW), approximately from equator to 35°S in the Subantarctic Mode Water (SAMW), and all latitudes in the Antarctic Intermediate Water (AAIW). Less ΔC_{anth} in the eastern basin than the western basin was caused by weaker ventilation driven by SAMW and AAIW formation and subduction and stronger Antarctic Bottom Water (AABW) formation in the former than the latter. In addition to the spatial heterogeneity, C_{anth} increase rates accelerated from the 1990s to the 2000s, consistent with the overall increase in air-sea CO_2 exchange in the South Atlantic Ocean.

1. Introduction

Oceanic uptake of atmospheric carbon dioxide (CO_2) is a key part of the global CO_2 budget. Human activities have increased the partial pressure of CO_2 (pCO_2) in the atmosphere from ~280 ppm before the Industrial Revolution to well above 400 ppm at present (Friedlingstein et al., 2020). The ocean has absorbed 25-30% of the anthropogenic CO_2 from the atmosphere and has played an essential role in regulating atmospheric CO_2 concentration and our planet's climate conditions (Sabine et al., 2004; Khatiwala et al., 2013; Le Quéré et al., 2016; DeVries et al., 2017; Gruber et al., 2019a; Friedlingstein et al., 2019). The absorbed CO_2 in surface waters is then transported to the interior ocean via ventilation, that is through mode, intermediate and deep-water formation. The uptake of atmospheric CO_2 has also increased seawater dissolved inorganic carbon (DIC) concentrations and decreased pH and carbonate mineral saturation (ocean acidification), with serious potential negative consequences on ocean ecosystem and biogeochemistry (Kleypas et al., 1999; Orr et al., 2005; Doney et al., 2009). Thus, monitoring the anthropogenic CO_2 or carbon (C_{anth}) uptake and storage or increase rate is important for climate science and marine ecosystem research. Key challenges include reducing the uncertainty in estimating C_{anth} via observations and modeling, as well as quantifying the spatial and decadal variations of the oceanic C_{anth} uptake rates (Doney, 2010).

The Meridional Overturning Circulation (MOC) plays a large role in the decadal variations of C_{anth} in the Atlantic, which contains approximately 38% of the total C_{anth} inventory although its volume is only 23% of the world ocean (Sabine et al., 2004). Overturning not only regulates the strength of ocean CO_2 uptake (Pérez et al., 2013), but is also critical for the ventilation of older water masses, facilitating uptake and storage of C_{anth} (Iudicone et al., 2011; Sallée et al., 2012). It has been suggested that decadal change of C_{anth} is regulated by the MOC variations in the ocean interior (DeVries et al., 2017). For example, Wanninkhof et al. (2010) reported that a decadal increase in C_{anth} inventory was anomalously higher in the South Atlantic Ocean compared to the North Atlantic during the 1990s (3.0 vs. 1.9 Pg C decade^{-1}). Woosley et al. (2016) observed a strengthened uptake rate of C_{anth} in both the South and North Atlantic Ocean basins during the 2000s (3.7-4.4 Pg C decade^{-1}). In the North Atlantic Ocean, the formation of deep water (North

Atlantic Deep water, NADW) and deep convection in the subpolar region represents the largest and deepest penetrated C_{anth} uptake regions (Sabine et al., 2004; Perez et al., 2018). However, the relatively shallow isopycnal surfaces associated with mode and intermediate waters are also important carriers of C_{anth}, as they are ventilated on time scales of a few decades or less. This ventilation on short timescales permits the transportation of C_{anth} from the outcrop regions at the mid to high latitudes rapidly toward the ocean's interior (Gruber et al., 2019b). An important region of oceanic C_{anth} uptake by this mechanism is in the subtropical and subantarctic South Atlantic Ocean, where mode (Subantarctic Mode Water, SAMW) and intermediate (Antarctic Intermediate Water, AAIW) waters form and carry CO_2 with them via subduction and lateral transport northward (McNeil et al., 2001; Sabine et al., 2004). Together with the deep convection process, this northward transport makes C_{anth}'s areal storage in the Atlantic twice that in the Pacific (Quay et al., 2007; DeVries et al., 2017).

The Mid-Atlantic Ridge separates the Atlantic Ocean into the western and eastern basin, with different geomorphology and circulation, and thus, C_{anth} uptake and storage rates may have both a meridional and zonal gradient. Although the C_{anth} in the entire Atlantic Ocean has been researched using different methods (Gruber, 1998; Wanninkhof et al., 1999; Lee et al., 2003; Vázquez et al., 2009; Peng and Wanninkhof, 2009; Wanninkhof et al., 2010; Woosley et al., 2016), past estimates in the South Atlantic mainly focus on the western basin (Ríos et al., 2010; Ríos et al., 2012; Salt et al., 2014). The few studies that have compared the two basins observed a higher C_{anth} in the western than the eastern basin (Lee et al., 2003; Murata et al. 2008). Ríos et al. (2012) also indicated that the southwestern Atlantic Ocean dominates the South Atlantic sink of C_{anth} in the western basin from 1972 to 2005. Although the above studies have directly or indirectly compared the differences between the two basins, the decadal C_{anth} change in the eastern South Atlantic basin and the zonal C_{anth} gradient on the two sides of the Mid-Atlantic Ridge are poorly understood. The differences between the decadal variability of oceanic C_{anth} uptake and storage rates estimates in the Atlantic Ocean on both meridional and longitudinal directions may be caused by the study time periods, methodological differences, spatial and temporary limitations of the field observations, differences in ocean dynamic processes, and limitations on quantifying ocean circulation changes.

The decadal C_{anth} uptake fluctuation in the South Atlantic is also closely linked to the air-sea CO_2 flux variations. The air-sea CO_2 flux varies substantially on different timescales, especially in the Southern Ocean (Landschützer et al., 2015, 2016; Gruber et al., 2019b). The Southern Ocean carbon sink weakened significantly during the 1990s (Le Quéré et al., 2007), but strengthened rapidly after reaching a minimum around the year 2000 (Landschützer et al., 2015). The reinvigoration during the 2000s brought the Southern Ocean sink strength back to the level expected from the increase in atmospheric CO_2 (Gruber et al., 2019b). These Southern Ocean fluctuations should affect CO_2 uptake rate, and thus the decadal variation of C_{anth} in the South Atlantic Ocean.

In this study, we used two meridional transects in the eastern (A13.5) and western (A16S) basin and one zonal transect (A10) to explore spatial and decadal variations of C_{anth} in the South Atlantic Ocean. First, we compared depth profiles of water properties to infer water mass distributions and transport, and the possible C_{anth} changes from physical and biogeochemical processes. Then, we constrained the distribution and variation of C_{anth} in the South Atlantic by comparing historical data from the 1980s to the more recent data in the 2010s. This allowed us to compare C_{anth} between the two sides of the Mid-Atlantic Ridge in the South Atlantic Ocean. Finally, we discussed C_{anth} distributions and changes in the context of physical oceanography (circulation of the mode and

intermediate waters) and air-sea fluxes in the eastern and western basins. A methodology study and a sensitivity test are given as an appendix.

2. Study Region, Sites, and Data

2.1 Description of Study Region and the Transects

The study region focuses on the South Atlantic Ocean from 60°W to 20°E (Figure 1a). The Mid-Atlantic Ridge separates the eastern and western Atlantic basins and rises to a depth of ~2000 m. Meridional transects A13.5 and A16S (Figure 1a) are located, on the east and west sides of the Mid-Atlantic Ridge, respectively. Transect A13.5, along the prime meridian, was occupied in 1983/1984 and 2010. Transect A16S, along roughly 25°W, was occupied in 1989, 2005, and 2013. In addition, a zonal section A10 along 30°S was occupied in 1992, 2003 and 2011.

The eastward flowing Antarctic Circumpolar Current (ACC) is the most prominent feature in the southern part of the South Atlantic Ocean. The ACC forms three main fronts: Southern ACC Front (SACCF), Polar Front (PF), and Subantarctic Front (SAF) (Figure 1a, Orsi et al., 1995). To the south of the SACCF is the Southern Boundary (SB, Figure 1a, Sievers & Emery, 1978), which is the southern edge of the low oxygen layer of the Upper Circumpolar Deep Water (UCDW). To the north of the SAF is the Subtropical Front (STF), which is the oceanographic northern boundary for the Southern Ocean (Talley, 2011). North of the ACC, the Brazil Current flows poleward near the western South Atlantic boundary (Evans et al., 2017; Bryden et al., 2011; Peterson & Stramma, 1991). Then it meets the cold and fresh northward Malvinas Current (Gordon & Greengrove, 1986). South America forces the Brazil-Malvinas Confluence offshore in a series of meanders, forming the eastward South Atlantic Currents. At the eastern South Atlantic boundary, the South Atlantic Currents turns northward to feed Benguela Current (Shillington, 1998; Evans et al., 2017). The Brazil Current, South Atlantic Current, and Benguela Current compose the anticyclonic South Atlantic subtropical gyre. On the north side of the subtropical gyre is the equatorial circulation in the tropical Atlantic. The tropical Atlantic is bisected by the Mid-Atlantic Ridge. The eastern tropical region is confined to the north by the African continent. The equatorial circulations are also shown in the map (Figure 1a).

The South Atlantic Ocean is a key component of the global MOC (Marshall & Speer, 2012). The MOC in the Atlantic (Atlantic Meridional Overturning Circulation, AMOC) is composed of a double cell (Figure 1b). The upper cell consists of the upper ocean waters which flows northward from the South Atlantic and becomes denser in the northern North Atlantic to feed the overturn in the North Atlantic; the overturn flows out southward at depth, eventually becoming North Atlantic Deep Water (NADW). The lower cell consists of the dense Antarctic Bottom Water (AABW) which flows northward and upwells into the lower part of the NADW, disappearing by the mid-latitude in the North Atlantic.

There are several important water masses in the South Atlantic (Figure 1b), including Surface Water (SW, which includes Antarctic Surface Water, Subantarctic Surface Water, and Subtropical Surface Water), SAMW, AAIW, UCDW, NADW, Lower Circumpolar Deep Water (LCDW), and AABW. SAMW is located in the upper layer (~ 0-600 m) between the surface water and intermediate water, formed from the thick winter mixed layers subducted north of SAF, which is distinguished by a potential vorticity minimum and an oxygen maximum (McCartney et al., 1977) in subtropical South Atlantic. The AAIW is located between the SAMW and the UCDW (~ 500-1200 m), which is characterized by a salinity minimum in subtropical South Atlantic and tropical Atlantic, and is formed from advection of fresh subantarctic surface water. NADW originates in the North Atlantic and penetrates into the South Atlantic between about 1500 and 3500 m depth

and has distinctively high salinity, high oxygen, and low nutrients. The southward NADW process is limited by interaction with the ACC. The UCDW is characterized by low oxygen and high nutrients. The LCDW includes the vertical salinity maximum derived from the NADW. The AABW forms along the coast of Antarctica, where very dense, cold near-surface shelf waters, created by ocean-atmosphere and ocean-ice interactions mix with upwelling LCDW (Doney & Hecht, 2002). The northward flowing AABW is characterized by an oxygen maxima, nutrient minima and cold waters in the bottom layer (~ 3000 m to bottom). (Rhein et al., 1998; Orsi et al., 1999).

Figure 1. (a) Map of South Atlantic. The red solid line represent position of A13.5 (2010), the red dashed line represent position of A16S (2013), and the red dotted line represent position of A10 (2011). The dark blue lines with arrows represents South Atlantic Ocean surface circulation schematics (Talley, 2011), Acronyms: North Equatorial Countercurrent (NECC), Northern branch of the South Equatorial Current (NSEC), Equatorial Undercurrent (EUC), Central branch of the South Equatorial Current (CSEC), South Equatorial Countercurrent (SECC), and the yellow lines represent fronts (Orsi et al., 1995), from south to north, Southern Boundary (SB), Southern ACC Front (SACCF), Polar Front (PF), Subantarctic Front (SAF), and Subtropical Front (STF); (b) water masses in the South Atlantic Ocean, and schematic diagram of the upper cell (white arrows) and the lower cell (dark arrows) of the Atlantic meridional overturning circulation (AMOC) in the South Atlantic, Water masses (along A16S) are: Surface Water (SW), Subantarctic Mode Water (SAMW), Antarctic Intermediate Water (AAIW), Upper Circumpolar Deep Water (UCDW), Lower Circumpolar Deep Water (LCDW), Antarctic Bottom Water (AABW). North Atlantic Deep Water (NADW) is located between UCDW and LCDW. Figure 1(b) is constructed based on A16S 2005 data.

2.2 Data

Data from the A13.5, A16S and A10 cruises (Table S1) were obtained from CCHDO (https://cchdo.ucsd.edu/) and GLODAPv2.2019 (https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2_2019/). The two datasets were matched using their EXPOCODEs. GLODAPv2.2019 provides primary and secondary quality control to remove outliers and correct for measurement biases (Olsen et al., 2019), which are listed in Table S1. In addition, all in-situ data and acceptable data (Quality flag=2) from A13.5, A16 and A10 hydrographic observations were counted. Overall, the number of acceptable total alkalinity (TA) and DIC samples were less than other variables, especially for the earlier cruises. In our study, temperature, salinity, neutral density, apparent oxygen utilization (AOU), nitrate, silicate, phosphate, and total alkalinity were considered as possible variables for our analysis. As no phosphate data is available for the A16S (2005) cruise in GLODAPv2.2019 dataset (cruise 343), data from 33RO20050111 (CCHDO) were used instead.

We also used sea surface pCO_2 and air-sea CO_2 flux from an observation- and neural network method-based global monthly gridded sea surface pCO_2 product from 1982 onward provided by Landschützer et al. (2020) (https://www.ncei.noaa.gov/access/ocean-carbon-data-system/oceans/SPCO2_1982_present_ETH_SOM_FFN.html). The spatial resolution is 1°x1°, covering the global ocean including the South Atlantic Ocean.

3. Methods

3.1 The eMLR Method

The extended multiple linear regression (eMLR) method was developed to take advantage of the reoccupation of the World Ocean Circulation Experiment / Joint Global Ocean Flux Study (WOCE/JGOFS) hydrographic sections. WOCE/JGOFS is now organized under the international Global Ocean Ship-based Hydrographic Investigations Program (GO-SHIP).

The repeat observations made it possible to obtain the DIC concentration differences between time t_1 and t_2: ΔDIC ($t_2 - t_1$). Wallace (1995) proposed a multiple linear regression (MLR) approach to estimate the anthropogenic carbon change between time t_1 and t_2: $\Delta C_{anth}(t_2 - t_1)$ using the difference in the two MLR fits, based on observations that marine inorganic carbon exhibits a strong correlation with other oceanographic variables over large areas of the ocean (Brewer, 1978; Chen & Millero, 1979). However, the estimated ΔC_{anth} is strongly biased, primarily created by an amplification of the residuals in the two fits. To overcome this shortage, Friis et al. (2005) introduced the extended MLR (eMLR) method to estimate $\Delta C_{anth}(t_2 - t_1)$ by subtracting the coefficients of the two fits. The eMLR approach removes a substantial fraction of the noise in the MLRs and tends to better separate the C_{anth} signal from the natural variations in DIC. Thus, the eMLR method has been widely used to determine the decadal increase in C_{anth} based on repeat hydrographic data (e.g., Friis et al., 2005; Sabine et al., 2008; Wanninkhof et al., 2010; Woosley et al., 2016; Carter et al., 2017). However, the eMLR method assumes that baseline correlations and the resulting residual fields will remain constant over time even under the effect of long-term climate changes. (Levine et al., 2008; Goodkin et al., 2011). Therefore, to obtain the reliable ΔC_{anth} (keep the error below 20%), the eMLR method should be used for cruise data collected within three decades (Clement & Gruber, 2018).

In this study, first, we carried out extensive method assessments based on statistical analysis and sensitivity study of choosing water masses and selecting parameters, and a careful comparison of several methods (see details in Appendix A and Supplementary documents). Finally, we adopted and applied a specific eMLR method to estimate the anthropogenic CO_2 change (ΔC_{anth}) for the water deeper than 50 m. Our method, described below, builds on the eMLR method (Friis et al., 2005) but includes several treatments to obtain optimal estimation of C_{anth} in the South Atlantic Ocean.

Step 1, water masses separation. Applying the regression equation to various isopycnal layers or characteristic water masses is a critical first step in the eMRL approach. In our study, we considered six typical water masses: SW, SAMW, AAIW, UCDW, LCDW, and AABW. They were divided into 5 layers based on neutral density (γ) (Table 1). Although other authors have found differences in C_{anth} increase rates between AABW and LCDW (Ríos et al. 2012), due to the smaller number of sampling points in AABW (neutral density>28.27 kg m^{-3}, Talley, 2011) and no sample's neutral density smaller than 28.27 kg m^{-3} in A13.5 (2010) cruise, the data in AABW and LCDW were combined into one layer (layer 5). NADW is transformed to Circumpolar Deep Water (CDW) in the South Atlantic Ocean (Santoso et al., 2006), and the density of NADW was similar to UCDW and LCDW, so we split it between layer 4 (with the UCDW) and layer 5 using neutral density 28 as a criterial. We have also compared our approach with other approaches such as dividing water column into 14 isoneutral slabs or 25 isopycnal layers (Appendix A).

Table 1. Neutral density limits for five layers and corresponding water mass. Definitions following Evans et al. (2017), Talley (2011) and Heywood & King (2002).

Layer	Neutral density (γ) limits	Water mass
1	γ< 26.80	Surface Water (SW)
2	26.80 <γ< 27.23	Subantarctic Mode Water (SAMW)
3	27.23 <γ< 27.5	Antarctic Intermediate Water (AAIW)

4	27.5<γ< 28	Upper Circumpolar Deep Water (UCDW)
	27.92<γ< 28	North Atlantic Deep Water (NADW)
5	28<γ< 28.11	North Atlantic Deep Water (NADW)
	28<γ< 28.27	Lower Circumpolar Deep Water (LCDW)
	γ >28.27	Antarctic Bottom Water (AABW)

Step 2, compute C*. Since the DIC changes in water masses are influenced by both natural and anthropogenic processes, it is desirable to introduce a new variable that removes natural change as much as possible. Assuming the stoichiometric ratios are constant, the influence of natural biological processes on the DIC can be removed by creating a new variable, C*, as described in Gruber et al. (1996) following the principle described earlier (Brewer, 1978; Chen & Millero, 1979). Since C* removes the carbon originating from biological processes, it captures air-sea gas exchange, including C_{anth} uptake (Clement & Gruber 2018). In a review paper, it was concluded that in most cases the differences between the C* approach and the original DIC-based Chen approach were rather small (Sabine and Tanhua, 2009). C* is computed from measured DIC, TA, and oxygen (Gruber et al., 1996):

$$C^* = DIC - r_{C:O_2}O_2 - \frac{1}{2}\left(\mathrm{TA} + r_{N:O_2}O_2\right) \quad (1)$$

where the stoichiometric ratios of P:N:C:O used in this study are 1:16:117:170 (Anderson & Sarmiento, 1994).

Step 3, choice of variables and combinations of regression. Several studies have shown that the results of the eMLR method are still sensitive to the selected predictors (i.e., Friis et al., 2005; Woosley et al., 2016). However, there are no universal criteria for which and how many variables should be used. Usually, variable selection is determined by assessments of the statistical fits, data availability, and data quality (Sabine et al. 2008). Salinity and temperature are used to characterize the physical properties in almost all research to date, while the majority of differences lie in the selection of nutrients (such as nitrate, silicate or phosphate), oxygen (or AOU), or TA to characterize the biological properties (Table S2). Here, we chose temperature (T, or potential temperature θ), salinity (S), neutral density (γ, or potential density σ), AOU, nitrate (N), silicate (Si), phosphate (P), and TA as possible predictors to establish the multiple linear regression (MLR) equation. The simple (Pearson) correlation coefficients between these variables were calculated to check the independence (see Additional Supporting Information: ASI_correlation_coefficients.xlsx). We selected 11 best combinations of regression according to multiple and partial correlation coefficients as well as Root Mean Square Error (RMSE) from more than 100 combinations. In previous studies, some prefer to choose temperature as a predictor, while others prefer to use potential temperature; a similar situation also exists in neutral density and potential density. In our work, both of temperature and potential temperature (neutral density and potential density) were used to test which one is better. We did not find much difference between these choices when using them as predictors in MLR (see Additional Supporting Information: ASI_correlation_coefficients.xlsx). The best choice of variables that can be used in the multiple linear regression function are given in Table 2. The mean ΔC_{anth} based on these combinations (Table 2) of predictors for the regressions were used for the final ΔC_{anth} (Carter et al. 2017, 2019; Gruber et al. 2019a).

Table 2. Predictors used in each of 11 regressions.

Regression. #	T (θ)	S	γ (σ)	AOU	N	Si	P	TA

1	✓	✓		✓	✓	✓			
2	✓	✓		✓		✓	✓		
3	✓	✓		✓	✓				
4	✓	✓		✓			✓		
5	✓	✓		✓		✓			
6		✓	✓	✓	✓	✓			
7		✓	✓	✓		✓	✓		
8	✓			✓	✓			✓	
9	✓			✓			✓	✓	
10			✓	✓	✓			✓	
11			✓	✓			✓	✓	

In addition, we used the "robust" multiple linear regression (Carter et al., 2017) instead of the "stepwise" regression (Wanninkhof et al., 2010; Woosley et al., 2016). Robust regression can eliminate outliers by iteratively re-estimating the regression coefficient after assigning a small weight to the measured value with large residual, so as to minimize the influence of outliers on the regression (Carter et al., 2017).

In the end, the general form of the equation fit to data is as follows:

$$C^*(t) = A + B \cdot P_1 + C \cdot P_2 + D \cdot P_3 + \cdots + Residuals \quad (2)$$

where A is the intercept, and B, C, D … etc. are the MLR coefficients, P_1, P_2, P_3, … P_n are selected predictor variables (e.g., T, S, γ, AOU, TA, and nutrients). And the residuals are the difference between the observed C* and the fitted one (Table 2). The part of changes in biology and physics are corrected by the multiple linear regression equations. Additionally, the C* further reduce the impact of biological processes. Then, the anthropogenic CO_2 change $\Delta C_{anth}(t_2 - t_1)$ was estimated by subtracting the coefficients of the two fits and selected predictor variables of the later observation:

$$\Delta C_{anth}(t_2 - t_1) = (A_2 - A_1) + (B_2 - B_1) \cdot P_1 + (C_2 - C_1) \cdot P_2 + (D_2 - D_1) \cdot P_3 + \cdots \quad (3)$$

As no TA data exists in the 1992 A10 survey, a slightly different variable, C^ ($C^{\wedge} = DIC - r_{C:O_2} O_2$, Carter et al., 2017) was used instead to establish the MLR for A10 (They are compared in the Appendix A). Additionally, since measurements of phosphate during the 2011 A10 survey were suspect from station 64 to 112 (~21°W-46°W), regressions with phosphate and TA were not used. Finally, the mean ΔC_{anth} from a combination of regression 1, 3, 5, 6 was used for the final ΔC_{anth} along A10 between 1992 and 2011. In order to make the ΔC_{anth} along A10 comparable in different time periods, we used the same treatments to estimate ΔC_{anth} along A10 between 1992 and 2003 and that between 2003 and 2011.

RMSE and residuals were used as eMLR error statistics, and thus helped to determine the best combinations and examine the uncertainty of the ΔC_{anth} estimation (see Additional Supporting Information: ASI_RMSE_residuals.docx). The mean RMSE in our MLR fits ranged from 2.9 to 4.3, with higher RMSE in SW and lower RMSE in the deep and bottom waters. The average residual of all 11 regressions for each cruise between the predicted and calculated C* based on observations mostly ranged from -0.69 to 0.22 μmol kg^{-1}. Our statistic errors were similar to or slightly improved over previous studies (Lee et al., 2003; Murata et al., 2008; Wanninkhof et al., 2010; Woosley et al., 2016).

3.2 ΔC_{anth} Estimation in the Mixed Layer

Past studies have shown that MLR and eMLR approaches are not suitable for the mixed layer waters (Sabine et al., 2008) for the following two reasons: they are strongly affected by seasonal

physical and biological processes and the exchange rate of CO_2 between seawater and atmospheric is relatively slow (~1 year). The depth of the mixed layer in the South Atlantic Ocean has a strong seasonal variation. The Mixed Layer Depth (MLD, Holte et al., 2017) is shallower in summer, and deeper in winter. The depth of summer mixed layer along A16S ranges from 16 meters to 61 meters and the depth along A13.5 ranges from 16 meters to 70 meters (Figure S1). The anthropogenic CO_2 change in the mixed layer (upper 50 m) was estimated based on Carter et al. (2017), assuming sea surface pCO_2 increase follows the atmospheric increase. We also confirmed that the sea surface pCO_2 increase rate is similar to the atmospheric pCO_2 increase rate in the South Atlantic Ocean (Figure S2, Table S3). The calculation had the following five steps: (1) calculate $pCO_2(t_2)$ of the later occupation based on TA(t_2), DIC(t_2), and carbonate constants (Dickson & Millero, 1987); (2) determine the increase in annual mean atmospheric pCO_2 (ΔpCO_2) between the two occupations according to the Mauna Loa observatory record (Keeling, 1986); (3) set $pCO_2(t_1) = pCO_2(t_2) - \Delta pCO_2$; (4) calculate DIC´($t_2$), using the TA($t_2$) and $pCO_2(t_1)$ assuming no further anthropogenic CO_2 uptake between the two occupations; (5) then, the anthropogenic CO_2 increase is equal to the difference between the measured DIC(t_2) and estimated DIC´(t_2) from step 4, or ΔC_{anth} = DIC(t_2) - DIC´(t_2). Note that despite its large uncertainty, ΔC_{anth} in the mixed layer is only a small part (<0.03%) of the total ΔC_{anth} inventory.

3.3 Column Inventory Change and Total Inventory Change of C_{anth}

The column inventory of ΔC_{anth} (μmol m^{-2}) was estimated by integrating the gridded ΔC_{anth} from the surface down to a specific depth, following Tanhua & Keeling (2012).

$$\text{Column inventory change} = \int_{surface}^{depth} \Delta C_{anth} \times rho \, dz \quad (4)$$

where ΔC_{anth} is the vertically interpolated anthropogenic carbon concentration change, rho is the density at in situ temperature and pressure.

Total inventory change of C_{anth} was calculated by averaging the column inventory change over meridional bands and multiplying by volume of water in that band (Woosley et al., 2016). The volumes of the South Atlantic Ocean can be found in Gruber et al. (1998) and Lee et al. (2003).

4. Results

4.1 Water Properties and their Decadal Changes

The spatial patterns of seawater properties are indicative of the large-scale water masses and their transport, and thus, they provide important context in explaining the spatial distributions of C_{anth} changes. The large-scale features along transect A16S were described in previous studies (e.g., Tsuchiya et al., 1994; Lee et al., 1997; Wanninkhof et al., 1999; Wanninkhof et al., 2010; Talley, 2011; Woosley et al. 2016) but the water properties in the eastern South Atlantic basin (along A13.5) have rarely been described. Here we compared the meridional depth profiles of water properties observed along A13.5 (2010) and A16S (2013). Salinity, AOU, nitrate, DIC, and CFC-12 were chosen to represent physical, biological and anthropogenic processes. Generally speaking, the main water property differences between transects A13.5 and A16S (Figure 2) were located in the mode and intermediate waters. Differences were also found in the deep and bottom waters (below 4000 m) in the tropical and subtropical region (north of 30°S). Similar results are also seen in the gridded differences between A16S and A13.5 in Figure S3, where the data were gridded on a 1° by 50 m grid with an inverse distance weighting (IDW) scheme (Wanninkhof et al., 2010) using a Matlab function.

Along both meridional transects, a low salinity tongue originated from surface at about 45-55°S, and then transported northward centered at about 1000 m and extended to the equator (Figure 2a & 2b). The low salinity region at the surface is indicative of the AAIW outcrop location, and the low salinity tongue represents AAIW, which is an important pathway for C_{anth} input into the ocean interior (Brewer, 1978). The fresh-water tongue in the intermediate water was more northerly along A16S than A13.5. The water mass was also fresher along A16S than A13.5 in the deep and bottom waters (north of 30°S and below 4000 m), that is, the low salinity waters in the AMOC lower cell extends northward along the bottom Cape Basin to as far as 30°S where it meets the Walvis Ridge.

AOU and nutrients are important indicators of biological processes. The highest AOU along both A16S and A13.5 were shown in the tropical region (around 10°S) at the subsurface layer (200-1200m) (Figure 2c & 2d). But the high subsurface AOU was higher and extended closer to the surface and further north along A13.5 (Figure 2d) than A16S (Figure 2c). Similar to AOU, nitrate (Figure 2e & 2f) was higher along A13.5 than A16S in the mode and intermediate waters, likely as a result of more accumulation of metabolic signals over slower and longer water transit time in the former. In addition, in the northeastern South Atlantic Ocean, a cyclonic upwelling region (Wacongne and Piton, 1992) was formed and had a large biological productivity, thus resulting in a large subsurface tropical oxygen minimum layer and nitrate maximum (Mercier et al., 2003). Finally, DIC showed close similarities with AOU and nitrate, because that the process of controlling AOU and nitrate also affected the distribution of DIC. DIC was higher in the subtropical intermediate water where AOU and nitrate had maxima. However, nitrate, AOU and DIC were lower along A13.5 than A16S in the bottom waters north of 30°S, indicating more northward water mass expansion in the latter.

Transient tracers such as the chlorofluorocarbons (CFCs) are good indicators of ocean physics and biogeochemistry processes (Fine, 2011). CFC-12 dissolve into the surface layer of the ocean and are subducted into the inner ocean during the outcrop events. It is a good tracer to trace anthropogenic signal from the atmosphere. CFC-12 was high at high latitude (48-60°S) near surface, and then it decreased with waters subducting downward and northward. Besides, high CFC-12 concentrations along both A16S (Figure 2i) and A13.5 (Figure 2j) were mainly shown in the upper 2000 m, which imply that C_{anth} absorbed from outcrop regions during outcrop events entered the upper ocean along the pathway of subduction. Combined with salinity pattern, new AAIW in the western basin also can be recognized near 45°S by a high CFC-12 tongue, spreading northward to approximately 35°S, then turning eastward and entering into subtropical gyre (Messias & Laurent, 1998). Consistent with the observed biogeochemical tracers, the CFC-12 signal was also stronger along A16S than A13.5 in the subsurface layer near the tropical region (south of 20°S). Finally, relatively high CFC-12 concentrations were observed in the AABW along A16S in particular near the Falkland Escarpment, indicating that the AABW spreads widely in the Argentine Basin (Mémery et al., 2000). In contrast, the A13.5 transect displays a weak CFC-12 distribution in the bottom. However, a slightly higher CFC-12 can be found near the equator at the bottom (Fig. 2j), implying bottom water and deep water across the Mid-Atlantic Ridge through the Romanche Fracture Zone (Arhan et al., 2003).

Figure 2. Vertical distribution of (a) & (b) salinity, (c) & (d) AOU, (e) & (f) nitrate, (g) & (h) DIC, and (i) & (j) CFC-12 along transect A16S (5°S-60°S) during 2013 Dec 23 – 2014 Feb 05 (left), and along transect A13.5 (5°N-55°S) during 2010 Mar 08 - Apr 18 (right). The

411 background shading in Figure 2i and 2j denote $\log_{10}$(CFC-12), while the contours represent the
412 values of CFC-12 concentration.
413
414 Decadal changes of water properties are also indicative of the physical and biogeochemical
415 variations over time, and they can be used to explain the temporary variations of C_{anth} changes.
416 The variation of water properties in the upper mixed layer is influenced by short-term seasonal
417 variability. Below the winter mixed layer, the water properties are mainly affected by changes in
418 circulation, ventilation, and remineralization of organic matter on annual to decadal time scales
419 (Wanninkhof et al., 2010; Woosley et al., 2016). Changes in the above water properties between
420 2010s and 1980s (about 25 years apart) for the transect A13.5 and A16S were analyzed by
421 subtracting the gridded data of the earlier cruises from the later ones (Figure S4). Significant
422 changes occurred in the tropical region within the surface and intermediate waters, which were in
423 good agreement with Wanninkhof et al. (2010) and Woosley et al. (2016). Among them, DIC
424 increased along most of the A16S (Figure S4g) and A13.5 (Figure S4h) transect, except at 1000-
425 2000 m from 5-30°S along the A13.5 transect. CFC-12 differences in the South Atlantic (Figure
426 S4i & S4j) showed the large-scale downward and equatorward penetration from high-latitude
427 regions. There were large increases in CFC-12 concentrations (~1 pmol kg^{-1} over 20 years), which
428 reached deeper depths near the subtropical gyre (25-45°S) due to SAMW and AAIW formation in
429 the South Atlantic Ocean. Relatively high CFC-12 differences between the two decades along
430 A13.5 (Figure S4j) were persistently closer to the surface compared with that along A16S (Figure
431 S4i), indicating deeper penetrate along the A16S.
432
433 4.2 Spatial distributions of ΔC_{anth}
434 Anthropogenic CO_2 uptake and storage increased along all three transects in the South Atlantic
435 Ocean over the period with two common features (Figure 3). First, high ΔC_{anth} was detectable in
436 surface and near surface waters. Second, ΔC_{anth} penetrated to deeper depths in the subantarctic and
437 subtropical oceans and was limited to shallower depths in the tropical oceans.
438 For the two meridional transects A16S and A13.5, the ΔC_{anth} spatial patterns reflected the
439 underlying sources from both air-sea interaction and water mass transport. High ΔC_{anth} (more than
440 10 μmol kg^{-1} $decade^{-1}$) extended from the surface down to a depth of 800 m from subantarctic
441 region (around 45°S) to tropical region (around 15°S) along A16S (1989-2013) (Figure 3a), which
442 is comparable with the ΔC_{anth} pattern in Woosley et al. (2016). In contrast, while ΔC_{anth} was also
443 high in the upper layer along A13.5 (1983-2010), it was lower than that along A16S (1989-2013)
444 in water masses below the SAMW. ΔC_{anth} along A13.5 from the surface down to 1000 m around
445 25-45°S was about 5～12 μmol kg^{-1} $decade^{-1}$ and was limited to the upper 400 m around the region
446 north of 18°S (Figure 3b), which is similar to the C_{anth} pattern along A14, another transect in the
447 eastern basin (Ríos et al., 2003). The meridional distribution pattern of the C_{anth} uptake and storage
448 changes in the eastern basin along A13.5 were much less in the tropical region and were shifted
449 more toward the subtropical and subantarctic regions. In contrast, in the western basin along A16S,
450 subsurface water in the subtropical and tropical regions had the highest ΔC_{anth}. The deepest and
451 largest C_{anth} increase occurred around 30-45°S in both sides of the Mid-Atlantic Ridge, but
452 penetration was deeper in the western basin than in the eastern basin during this period.
453 Below 2000 m, ΔC_{anth} was low along A13.5 (~ 1 μmol kg^{-1} $decade^{-1}$, Figure 3b). In contrast, a
454 high ΔC_{anth} signal of ~ 3 μmol kg^{-1} $decade^{-1}$ was clearly seen in the LCDW and AABW along
455 A16S (Figure 3a). Wanninkhof et al. (2010) also observed an increase in ΔC_{anth} of 3 to 4 μmol kg^{-1}
456 in the AABW along A16S from 1989 to 2005. The increased ΔC_{anth} in the deep and bottom waters

was likely a result of the deep ventilation pathway, a conclusion supported by the finding of relatively high CFC-12 concentrations there. Ríos et al. (2012) also reported that the AABW in the western basin of the South Atlantic contained higher ΔC_{anth} than that in the upper layer of AABW (1.5 vs. 0.8 μmol kg^{-1} $decade^{-1}$). In our study, CFC-12 (Figure 2i & 2j) was used as an evidence to support the transport of surface water into the ocean interior and support a higher ΔC_{anth} in the AABW along A16S than A13.5.

The zonal transect (A10) was used to further examine the west-east differences in C_{anth} change here as it crosses both A16S and A13.5 and has been used in the past to provide a basin-wide spatial variation of ΔC_{anth} (i.e., Murata et al., 2008; Woosley et al., 2016). Vertically, ΔC_{anth} along A10 between 1992 and 2011 was high in the upper 1000 m depth layer compared to the deeper depths (Figure 3c). Meanwhile, the ΔC_{anth} was slightly (about 1 μmol kg^{-1} $decade^{-1}$) higher west of 20°W than east of 20°W in the upper 500 m. However, the distribution of ΔC_{anth} was quite uniform from west to east below 500 m, except a lower ΔC_{anth} towards the eastern end. The nearly parallel isopycnals suggested C_{anth} within the water mass was transported along the isopycnal layers and was not ventilated. A10 crosses A16S and A13.5 along the 30°S at 25°W and 0°, respectively (the dashed lines in Figure 3). The ΔC_{anth} values at the crossover locations were not greatly different. Therefore, our work suggests that transect A10 only minimally reflects the zonal spatial differences of the South Atlantic Ocean. The eastern versus western basin contrast will be better reflected by a complete comparison of A13.5 to A16S at all latitude bands.

Figure 3. Anthropogenic CO_2 changes along (a) A16S (1989-2013), (b) A13.5 (1983/84-2010) and (c) A10 (1992-2011). The white lines in Figure 3a, 3b and 3c depict neutral density isopycnals along A16S (2013), A13.5 (2010) and A10 (2011), and separate characteristic water mass layers. The gray dashed line in Figure 3a and 3b indicates the latitude of zonal transect A10, while the gray dashed lines in Figure 3c indicate the longitudes of meridional transect A16S and A13.5.

4.3 Decadal differences in ΔC_{anth}

The above A16S and A10 calculations of ΔC_{anth} used the differences between 1989 and 2013 and 1992 and 2011, respectively (Figure 3). We also used two midpoint cruises to explore ΔC_{anth} on shorter time scales and observe possible changes between the earlier and later periods. However, there was no mid-point observation for A13.5. Overall, ΔC_{anth} was higher in the more recent time period compared to the older one. For transect A16S, ΔC_{anth} increased significantly in all water masses except for NADW for 2005-2013 (Figure 4a) compared to 1989-2005 (Figure 4b). The increase was largest (> 5 μmol kg^{-1} $decade^{-1}$) north of 50°S and in the upper 800 m. The highest ΔC_{anth} along A16S (2005-2013) was up to 15 μmol kg^{-1} $decade^{-1}$ in the upper 400 m approximately from 45°S to 15°S, and high ΔC_{anth} (~ 8～12 μmol kg^{-1} $decade^{-1}$) penetrated roughly along 27.23 kg m^{-3} isopycnal (boundary between the SAMW and the AAIW) from the surface near 50°S down to 1000 m near 25°S. However, high ΔC_{anth} (~ 10 μmol kg^{-1} $decade^{-1}$) along A16S (1989-2005) was mainly located north of 42°S and in the upper 500 m. For ΔC_{anth} along A10, the increase from 1992-2003 to 2003-2011 was mostly in the upper 700 m (Figure 4c and 4d). A small west-east difference existed along A10 (2003-2011), with a nearly constant change to a depth of 700 m. ΔC_{anth} along A10 (1992-2003) showed a different pattern in the upper 500 m, where the west part clearly contained more ΔC_{anth} than the east. Interestingly, the high ΔC_{anth} along A10

(2003-2011) was confined within the SW and SAMW (<27.23 kg m^{-3} isopycnal) but ΔC_{anth} along A10 (1992-2003) extended into the AAIW (<27.5 kg m^{-3} isopycnal).

Figure 4. Anthropogenic CO_2 changes along (a) A16S (2005-2013), (b) A16S (1989-2005), (c) A10 (2003-2011) and (d) A10 (1992-2003). The gray dashed line in Figure 4a and 4b indicates the latitude of zonal transect A10, while the gray dashed lines in Figure 4c and 4d indicate the longitudes of meridional transect A16S. The white lines depict neutral density isopycnals and separate characteristic water mass layers.

4.4 Column Inventory Changes Along Transects

Water column inventory of anthropogenic CO_2 increase is an important quantity reflecting the total amount of ΔC_{anth} in a region from both local atmospheric input and net lateral transport. Since changes in C_{anth} were mainly in the top 2000 m, here we focus on the column inventory changes in the upper 1000 m and 2000 m layers (Figure 5). Column inventory changes from the surface to the bottom (~ 6000 m) are given in Figure S5. The following features emerged from this analysis: (1) ΔC_{anth} column inventory was substantially higher along the A16S than A13.5 (red vs. black lines). (2) There were clear meridional variations in the ΔC_{anth} inventory distribution with the highest water column inventory change in the subantarctic and subtropical regions. (3) Decadal ΔC_{anth} inventory increases accelerated from the 1990s to the 2000s.

The column inventory changes of C_{anth} from the 1980s to 2010s showed a similar meridional pattern for A16S and A13.5, (Figure 5a, red lines and black lines, respectively), which increased from high latitude (~ from 55°S to 40°S), peaked at 30-40°S, then decreased to the equator. The mean upper 2000 m column inventory change was 0.91±0.25 mol m^{-2} yr^{-1} and 0.57±0.22 mol m^{-2} yr^{-1}, respectively, for A16S (2005-2013) and A13.5 (1983-2010). The column inventory change was about 0.34±0.15 mol m^{-2} yr^{-1} higher in the western basin (A16S) than the eastern basin (A13.5) within the upper 2000 m. The largest west-east difference occurred at low latitudes, and then gradually decreased toward higher latitudes (Figure 5a). However, the differences in the column inventory changes in the upper 1000 m along the two transects were relatively small, especially in the region south of 25°S (Figure 5a, red dashed lines vs. black dashed lines). Thus, A16S contained more ΔC_{anth} than A13.5 mostly in the depth between 1000 and 2000 m. For the zonal transect A10 (1992-2011), the mean column inventory change (0-2000 m) was 1.04±0.15 mol m^{-2} yr^{-1}, as it crosses roughly the highest locations of ΔC_{anth} in both A16S and A13.5. More importantly, C_{anth} column inventory change along A10 showed only a very limited west-east contrast.

The annual column inventory change rate along A16S between 2013 and 2005 (Figure 5a) was much higher than that between 2005 and 1989, in agreement with an increase in the atmospheric CO_2 growth rate (https://gml.noaa.gov/ccgg/trends/gr.html). During both periods, the largest increases in annual C_{anth} column inventory changes (about 0.78 mol m^{-2} yr^{-1}) were located at mid-high latitudes (30-50°S), then the increase gradually decreased towards higher or lower latitudes (Figure 5a, Table S4). Wanninkhof et al., (2010) estimated an average uptake rate of 0.76±0.2 mol m^{-2} yr^{-1} for A16S (south of 15°S during 1989-2005), which is in good agreement with our average of 0.76±0.25 mol m^{-2} yr^{-1} for the top 2000 m (Figure 5a, green solid line) during the same time period. The mean C_{anth} column inventory change in the upper 2000 m for A16S (2013-2005) was 1.24±0.37 mol m^{-2} yr^{-1}, which was higher than the previous decade. Similarly, the ΔC_{anth} column inventory along the zonal transect A10 went through the parallel decadal variation. The column inventory change rate along A10 was higher during the 2000s (2003-2011) than the 1990s (1992-2003). The ΔC_{anth} difference along A10 between the two periods were mainly located in the

western part (west of 20°W) for the top 2000 m (Figure 5b, blue dotted line vs. green dotted line). Murata et al. (2008) estimated an average uptake rate of 0.6±0.1 mol m^{-2} yr^{-1} for the entire section A10 between 1992/1993 and 2003, while the average rate between 2003 and 2011 estimated by Woosley et al. (2016) is 0.83±0.1 mol m^{-2} yr^{-1}. Our values in the upper 2000 m are 1.11±0.22 mol m^{-2} yr^{-1} (2003-2011) and 0.99±0.12 mol m^{-2} yr^{-1} (1992-2003), which are somewhat larger than the values from these two studies.

Figure 5. Column inventory of ΔC_{anth} along (a) A16S and A13.5, (b) A10 for each time period in the top 2000 m and 1000 m.(c) Mean column inventory change for the top 2000 m as observed from A16S, A13.5 and A10 during different time periods, standard deviation is shown as vertical bars with matching colors along the right axes. Note that small column inventory changes around 55°S along A16S in Figure 5a is due to shallow bathymetry.

5. Discussion

5.1 Basin-wide Total Inventory Change in the South Atlantic Ocean

The total inventory of ΔC_{anth} in the Atlantic Ocean have been reported in previous studies, mostly based on combined Atlantic database during their study periods (e.g. Gruber, 1998; Lee et al., 2003; Touratier & Goyet, 2004; Vázquez-Rodríguez et al., 2009; Peng & Wanninkhof 2010) or repeat transects of A16 (Wanninkhof et al., 2010; Woosley et al., 2016). However, to our knowledge, there is no previous study published on decadal C_{anth} change for the entire repeat occupations of A13.5 and the basin wide inventory based on both A16S and A13.5.

Extrapolating column inventory changes from one or a few sections to the whole basin may lead to significant uncertainty, because of limited observations, spatial-temporal variation of ΔC_{anth}, and methodological challenges. Earlier, it was suggested that A16 (both the north and south transect) could be used to provide a good estimate for the entire Atlantic Ocean from the Arctic to the Antarctic (Wanninkhof et al., 2010; Talley, 2011). However, there are significant west-east ΔC_{anth} variability in the South Atlantic Ocean as is demonstrated here. Thus, A16S may not be representative for the entire South Atlantic Ocean. To evaluate this issue, we used both A16S (western basin) and A13.5 (eastern basin) and compared this to that using only A16S (Table 3). The total C_{anth} uptake in the South Atlantic Ocean (0-6000 m) from the 1980s to the 2010s estimated from the sum of the western basin and eastern basin was 27% lower than that based on only A16S (3.86 vs. 5.27 Pg C $decade^{-1}$). Thus, past publications may have overestimated the total C_{anth} uptake rate based on A16S only in the South Atlantic Ocean. Moreover, our total inventory change value estimated by both A16S and A13.5 in the South Atlantic Ocean is very close to the most recent value provided by Gruber et al. (2019) (3.86 Pg C $decade^{-1}$ between the 1980s and the 2010s vs. 5.9 Pg C between 1994 and 2007 or 4.2 Pg C $decade^{-1}$). It is also demonstrated that using both A16S and A13.5 to represent the total inventory in the South Atlantic Ocean is better than using A16S solely.

Note if we use only A16S to represent the entire basin, it appears that we would derive a total inventory change higher than that of Woosley et al. (2016) (e.g., 7.02 vs. 4.0 Pg C $decade^{-1}$ between 2005 and 2013) during the same period (Table S5). This is mainly because our column integration (0-6000 m) included ΔC_{anth} in the deep and bottom waters, which have ΔC_{anth} of about 3 μmol kg^{-1} $decade^{-1}$, while their integration stopped at ΔC_{anth} = 3 μmol kg^{-1}. This choice results in our column inventory change to be about 0.7 mol m^{-2} yr^{-1} higher than that integrated from the surface down to 2000 m (Figure S5, Table S4). If we use the mean column inventory along A16S

in the upper 2000 m to estimate the total inventory change, we get a total inventory change in the South Atlantic Ocean very close to that of Woosley et al. (2016) (4.0 vs. 4.0 Pg C decade^{-1}).

Table 3. Total inventory changes of C_{anth} (Pg C decade^{-1}) (0-6000 m) for each 10° latitude band in the South Atlantic Ocean between 1980s and 2010s.

Latitude band	Total Inventory Change			
	West (A16S)	East (A13.5)	Summary	Use only A16S [a]
0°S - 10°S	0.40±0.01	0.16±0.002	0.56±0.01	0.87±0.02
10°S - 20°S	0.47±0.02	0.12±0.006	0.59±0.02	0.90±0.05
20°S - 30°S	0.52±0.03	0.13±0.007	0.65±0.03	1.00±0.05
30°S - 40°S	0.54±0.02	0.30±0.012	0.84±0.02	1.04±0.04
40°S - 50°S	0.63±0.03	0.21±0.034	0.84±0.04	1.03±0.05
50°S - 60°S	0.31±0.13	0.06±0.007	0.37±0.13	0.43±0.18
Total South Atlantic	2.87±0.14	0.99+0.03	3.86±0.14	5.27±0.20

Note. [a]In the last column, A16S-based western basin results are extrapolated to the entire South Atlantic Ocean.

Finally, our results show that the decadal inventory changes over 10° latitude bands indicated a meridional pattern, similar to that reported by Wanninkhof et al. (2010) and Woosley et al. (2016) (Table 3). This meridional pattern was also in accord with the pattern of total inventory from Touratier & Goyet (2004), Lee et al. (2003), and Gruber (1998). Higher decadal inventory changes occurred in the middle latitudes (20-50°S), whereas they were minor in the low (0-20°S) and high latitudes (50-60°S). The column inventory changes over 10° latitude bands between A16S and A13.5 (Table S4) showed that the smallest difference was at 50-60°S and the largest difference was at 10-20°S. However, the total inventory changes over 10° latitude bands between western and eastern basins (Table 3) showed that the largest difference at 40-50°S, followed by 20-30°S, partly influenced by the volume difference in the eastern and western basins (Table S5). The total inventory changes in the western South Atlantic basin increased for all six latitude bands from the 1990s to the 2000s (Table S5). The largest increase occurred between 40-50°S, followed by that between 30 and 40°S. The decadal inventory changes had larger uncertainty at high latitudes.

5.2 Spatial Difference of ΔC_{anth} in Characteristic Water Masses

In the South Atlantic Ocean, the greatest ΔC_{anth} was observed in the surface and upper layers, consistent with the expectation since anthropogenic CO_2 came from the sea surface exchange with the atmosphere. There exists an upwelling region caused by strong westerlies south of the PF (around 60°S; Marshall & Speer, 2012) in the South Atlantic Ocean (Figure S6). The upwelled water with high DIC (Figure 2g & 2h) reaches the surface, is modified by biology, before being transported northward and subducted along AAIW and SAMW. The northward waters, which are part of the AMOC upper cell (Figure 1b), are exposed to the atmosphere in a region with high wind speeds (Figure S6), allowing for quicker air-sea CO_2 exchange (Figure S7), leading to higher uptake fluxes and absorption of C_{anth}. Therefore, high ΔC_{anth} is perceptible in the surface and upper layers due to oceanic CO_2 uptake at the air-sea interface and transport into ocean interior associated with the formation and subduction of SAMW and AAIW. However, there were regional differences in ΔC_{anth} on the two sides of the Mid-Atlantic Ridge, and ΔC_{anth} differences along A13.5 and A16S varied differently in each layer (Figure 3).

In the SW, the mean ΔC_{anth} values of both A16S and A13.5 were similar and mainly clustered around 10 μmol kg^{-1} decade^{-1} in the subantarctic and subtropical region (~20-40°S) (Figure 6a).

This is firstly because of the active coupling of C_{anth} to air-sea CO_2 exchange, and secondly our assumption of the sea surface pCO_2 increase following the atmospheric increase to estimate ΔC_{anth} in the mixed layer. However, air-sea CO_2 flux along A16S (Figure 7a) is about 1 mol m^{-2} yr^{-1} stronger than that along A13.5 (Figure 7b) south of 30°S, that is, the western basin takes up more CO_2 than the eastern basin in this region (Figure S7). What is more important is that C_{anth} absorbed by surface water does not stay for a long time but is then transported into ocean interior by ventilation, resulting in the similar ΔC_{anth} values in the surface water. In addition, the air-sea carbon flux along A16 and A13.5 experienced the same fluctuation from 1980s to 2010s.

The most significant ΔC_{anth} differences along A16S and A13.5 appeared in the SAMW and AAIW. In the SAMW, ΔC_{anth} was higher from 35°S to the equator along A16S than that along A13.5 (7.06±1.05 vs. 4.44±2.70 μmol kg^{-1} $decade^{-1}$ in Figure 6c). The largest ΔC_{anth} difference between the two transects occurred nears 20°S, with the west side having ΔC_{anth} more than twice of that of the east side. The ΔC_{anth} difference in the AAIW (Figure 6e) between A13.5 and A16S was evident at almost all latitudes (north of 50°S). Average ΔC_{anth} was 2.34±2.03 μmol kg^{-1} $decade^{-1}$ along A13.5 and 5.49±1.12 μmol kg^{-1} $decade^{-1}$ along A16S. The SAMW is formed by deep winter convection around the SAF (Figure 1b). It initially moves with the Malvinas Current (Figure 1a) and subducts northward. At the same time, this thick and homogeneous SAMW is advected eastward along the SAF into the subtropical gyre (Figure 1a). The AAIW is originated in the Drake Passage region (50-60°S, Figure 1a), formed through the Ekman transport process and the divergence and convergence of water masses. AAIW is advected eastward away from the Malvinas-Brazil Current confluence. Then after entering the South Atlantic Current (Figure 1a), it plunges downward to just beneath the thermocline in the subtropical South Atlantic. Although SAMW and AAIW subduct in the entire basin, the mean subduction is stronger in the western basin than eastern basin (Sallée et al., 2010). Thus, a relatively large amount of surface water with more C_{anth} is transported to the subtropical pycnocline, resulting in larger C_{anth} in the western basin. The depth and thickness of SAMW in the South Atlantic western basin are larger than those in eastern basin (Feucher et al., 2019, Figure S8a and S8b), which also implies mode water is dominated in the western basin. And a low PV along A16S (Figure S8c) ranges from 30°S to 40°S at the depth of 180-400 m implies the core of SAMW along A16S, which does not exist along A13.5 (Figure S8d), also indicating that mode water is obviously in the western basin. The fact that SAMW and AAIW advect eastward along the SAF and ventilate more recently in the western basin (Murata et al., 2008), implies that these water masses are younger in the western basin than eastern basin (see Figure 2i & 2j, where CFC-12 concentration along A16S is higher than A13.5). In addition, cumulative transport cross 35°S also illustrate that transports in the west part are larger than that in the east part (Dong et al., 2009). As a result, more C_{anth} must be transported northward in the western basin. In summary, mode and intermediate water in the western basin contain more recent and higher C_{anth} than that in the eastern basin.

ΔC_{anth} was also much lower along A13.5 than A16S in the tropical region, especially in the SW, SAMW and AAIW. This is because mode water and intermediate water carrying C_{anth} cannot effectively move northward, due to the blockage of water mass transport by the central African continent. The low salinity tongue (Figure 2a & 2b) also implies water mass transport less northward along A13.5 than A16S. Thus, the rate of northward movement of the subsurface waters in the eastern South Atlantic basin is slower than that in the western South Atlantic basin, resulting in less accumulation of C_{anth}. This slower water mass movement is also consistent with more accumulated biological signals such as higher AOU and nitrate maxima in the eastern part of SAMW and AAIW (Figure 2d & 2f). This is also consistent with the CFC-12 distribution (Figure

2i & 2j). The CFC-12 signal was lower along A13.5 than A16S in mode and intermediate waters in the subtropical region, indicating older waters (Figure 2j). In addition, the current system affected by the local topography plays an essential role in regulating the distribution of C_{anth}. For instance, the upwelled waters at the Angola Dome with large biological productivity (Talley, 2011) also have consumed CO_2, probably resulting in the low C_{anth} in this region. The presence of the southward flowing western boundary current enables the water and its C_{anth} signal to penetrate deeper in the western basin compared to the eastern half of the basin in the South Atlantic Ocean (Wanninkhof et al., 2010; Ríos et al., 2010; Vázquez-Rodríguez et al., 2009).

In the UCDW, ΔC_{anth} ranged from 2.55±1.09 to 3.89±0.95 μmol kg^{-1} decade^{-1} along A16S, while it was lower and varied from 1.00±0.38 to 2.46±1.11 μmol kg^{-1} decade^{-1} along A13.5 (Figure 6g). ΔC_{anth} was about 1-1.7 μmol kg^{-1} decade^{-1} higher along A16S than A13.5 over the entire transect from south to north in LCDW and AABW (Figure 6i). The existence of a substantial amount of ΔC_{anth} in the deep and bottom waters (UCDW, LCDW and AABW) along A16S but likely not along A13.5 is also consistent with CFC distributions along the two transects. It appears that in the South Atlantic, a major location of forming the densest Antarctic waters from brine rejection is in the Weddell Sea, leading to C_{anth} from air-sea CO_2 exchange sinks into the bottom water through vertical mixing. The deep gap in the South Scotia Ridge allows AABW with C_{anth} to flow northward. When it crosses the ACC, it extends north into the western South Atlantic, reaching the Brazil Basin (Talley, 2011; Abrahamsen et al., 2019). But the AABW cannot readily enter into the eastern South Atlantic basin due to the complex topography. Only a small amount of AABWs can eventually enter into the Cape basin through the Namib Col in the Walvis Ridge along southeastward flow and transport across the Mid-Atlantic Ridge though the Romanche Fracture at the equator along the eastward flow (Hogg & Thurnherr, 2005). Consequently, the western South Atlantic has a deep anthropogenic CO_2 invasion below 2000 m while this process hardly happens in the eastern basin.

In summary, more C_{anth} was transported to depth through the subduction of mode and intermediate waters in the western basin than the eastern basin, due to a stronger ventilation in the western basin than the eastern basin and the difference in circulation transport. ΔC_{anth} was also slightly higher (~1-2 μmol kg^{-1} decade^{-1}) in the deep and bottom waters of the western basin than that of the eastern basin, due to topographic limits that prevent the bottom water containing C_{anth} enter into the eastern basin.

Figure 6. The mean decadal anthropogenic CO_2 change (±1 Standard Deviation) along transect A13.5 and A16S (left), and A10 (right) over 5 latitude bands for each layer, (a) & (b) SW, (c) & (d) SAMW, (e) & (f) AAIW, (g) & (h) UCDW, and (i) & (j) LCDW+AABW.

Figure 7. Time-series of air-sea CO_2 flux along (a) A16S, (b) A13.5 and (c) A10. * Monthly gridded sea surface pCO_2 product (Landschützer et al., 2020) was matched by the longitude and latitude for each cruise. Positive air-sea CO_2 flux means ocean is carbon source, negative air-sea CO_2 flux means ocean is carbon sink.

5.3 ΔC_{anth} Increase Accelerated from the 1990s to 2000s

In addition to the spatial difference between the two basins, data from the multiple cruises along A16S and A10 also revealed decadal variations. Past studies have shown substantial decadal

variations in the anthropogenic CO_2 uptake rates in various parts of the world ocean (i.e., Sabine et al., 2008; Wanninkhof et al., 2010; Woosley et al., 2016; Carter et al., 2017).

The decadal C_{anth} increase or ΔC_{anth} accelerated from 1990s to 2000s along both A16S and A10. As a result, the mean column inventory change increased along A16S for the top 2000 m from 0.76 mol m^{-2} yr^{-1} during 1989-2005 period to 1.24 mol m^{-2} yr^{-1} during 2005-2013 period (Figure 5a & 5c). The mean column inventory change also increased slightly along A10 from 1992-2003 to 2003-2011 (Figure 5b & 5c). Decadal acceleration of ΔC_{anth} was in a good agreement with the atmospheric CO_2 increase rate and air-sea CO_2 flux variations. Atmospheric CO_2 increased from 353.1 ppm in 1989 to 379.8 ppm in 2005 and 396.52 ppm in 2013, resulting in the mean increased rate increased from 1.68 ppm yr^{-1} during 1989-2005 period to 2.11 ppm yr^{-1} during 2005-2013 period. Net air-sea CO_2 exchange affects the oceanic CO_2 absorption in the ocean surface layer, and via the subsequent transport in the ocean, will directly affect the basin-wide oceanic ΔC_{anth} changes.

Vertically, decadal ΔC_{anth} trends varied differently between water masses. In the SW, decadal ΔC_{anth} along both A16S and A10 increased significantly from the previous to the later decade (Figure 6a and 6b). This accelerated increase was supported by the rate of CO_2 uptake from the air-sea interface as SW was the most active layer of air-sea exchange. Below the SW, decadal ΔC_{anth} along A16S (Figure 6c, 6e, 6g & 6i) in the 2000s was also higher than that in 1990s in the subantarctic region (~25-50°S), but the decadal changes were more similar in the tropical region (north of 20°S). Importantly, as the ocean took up more CO_2 in the 2000s near the outcrop regions, the water masses then subducted and transported via mode and intermediate water formation carrying higher CO_2 into the inner ocean. This transport also was observed by the CFC-12 differences (Figure S4). The outcrop regions around 45°S took up large amounts of CO_2 before it subducted through mode and intermediate waters. As the subduction and northward transport rate was high in the region approximately from 45°S to 20°S (Feucher et al., 2019), and it slowed down at about 20°S, the decadal ΔC_{anth} acceleration along A16S from the 1990s to the 2000s was most clear south of 20°S (Figure 6).

6. Conclusions

Decadal anthropogenic CO_2 changes in the South Atlantic Ocean were assessed with a specific eMLR method using data from 1980s to 2010s along two meridional transects A13.5 and A16S and one zonal transect A10. An anthropogenic CO_2 increase of 3.86±0.14 Pg C $decade^{-1}$ was derived using column inventory changes along A16S and A13.5 extrapolated to the entire South Atlantic basin (0-60°S), which was consistent with past studies. The general pattern of significant increase in ΔC_{anth} appeared in SW, SAMW and AAIW above 27.23 kg m^{-3} neutral density contour line, caused by deep penetration associated with the formation and subduction of mode and intermediate waters. Our ΔC_{anth} along both A16S and A13.5 showed that the penetration and northward transport occurred around 25-50°S. The deepest penetration depth of ΔC_{anth} mainly reached 1000 m near 40°S along A16S, and 800 m near 32°S along A13.5, respectively.

Comparison of the transects A16S and A13.5 revealed large west-east differences: C_{anth} uptake rate in the top 2000 m along A16S (0.91±0.25 mol m^{-2} yr^{-1}, from 6°S to 60°S) was almost twice that along A13.5 (0.57±0.22 mol m^{-2} yr^{-1}, from 5°N to 54°S). The ΔC_{anth} column inventory change difference between A16S and A13.5 was largest at 10-20°S, and then gradually decreased to almost zero toward higher latitudes. ΔC_{anth} along A10 at 30°S also revealed west-east differences but the differences were generally smaller than those at the other latitudinal bands.

Vertical structure of the ΔC_{anth} between the two sides of the Mid-Atlantic Ridge also showed large differences in different water mass layers. The ΔC_{anth} difference (A16S minus A13.5) was about 3 μmol kg^{-1} $decade^{-1}$ north of 20°S in the SW, from 35°S to the equator in the SAMW and almost over all latitudes in the AAIW. For the deep and bottom waters (UCDW, LCDW, and AABW), ΔC_{anth} was about 1.6 μmol kg^{-1} $decade^{-1}$ higher along A16S than A13.5 for all latitudes. Differences in northward transport and recent outcrop events due to the formation of SAMW and AAIW between the western and the eastern basins were the main reasons for the ΔC_{anth} differences in SAMW and AAIW. While differences in topography accounted for deep water circulation causing the ΔC_{anth} differences in the deep and bottom waters.

ΔC_{anth} also increased by ~0.48 mol m^{-2} yr^{-1} from 1989-2005 to 2005-2013 along A16S, and by ~0.12 mol m^{-2} yr^{-1} from 1992-2003 to 2003-2011 along A10 in the top 2000 m. ΔC_{anth} accelerated for all latitudes along A16S and for all longitudes along A10 in the SW. For the meridional transect A16S, the highest ΔC_{anth} acceleration occurred at mid-high latitudes (south of 20°S) below the SW. While for the zonal transect A10, the highest ΔC_{anth} acceleration occurred in the east part (east of 25°W) in the SAMW, AAIW and UCDW. Increased CO_2 uptake from atmosphere and the subsequent northward transport via mode and intermediate water formation were the main reasons account for the ΔC_{anth} acceleration in the South Atlantic Ocean.

Appendix A: eMLR Sensitivity Tests and Method Comparisons

Using the strong correlation between carbon and other oceanographic parameters (Brewer, 1978; Chen & Millero, 1979) and the repeat occupations of the hydrographic sections, the MLR method was proposed by Wallace (1995) to estimate ΔC_{anth}. Since then, many studies have made modifications and improvements based on this method, in an attempt to improve the reliability and accuracy of ΔC_{anth} estimation. One of the most successful and recognized is eMLR approach proposed by Friis et al. (2005), which cancels a portion of carbon fit error. However, there are extensive comparisons and debates in the literature on using various approaches to estimate C_{anth} changes (Sabine et al., 2008; Wanninkhof et al., 2010; Williams et al., 2015; Quay et al., 2017; Carter et al., 2017, 2019; Clement and Gruber, 2018). The differences are mainly in the following three aspects: 1) how the data are divided into subsets or fit along isopycnal intervals; 2) how biological processes are corrected, and 3) how the predictors are selected. For example, Sabine et al. (2008) divided the cruises into different regions, Wanninkhof et al. (2010), Quay et al. (2017) and Clement and Gruber (2018) applied eMLR approach along isopycnal intervals, and Carter et al. (2017) used a moving window to separate data into subsets. To correct biological processes, Sabine et al. (2008) and Carter et al. (2017) made an adjustment based on AOU changes, Clement and Gruber (2018) used the C* as the dependent variable, and Carter et al. (2019) used the C^ instead.

Estimating C_{anth} using the same dataset but different methods could lead to different results. Here, we carried out a sensitivity test for the water mass identification used in the eMLR method and compared the specific eMLR approach presented here with alternative methods. Selecting the appropriate water masses or isopycnal intervals is a critical step in the effectiveness of the eMLR method. In this work, we built 11 regression equations for 5 layers containing six characteristic water masses (SW, SAMW, AAIW, UCDW, and the combined LCDW and AABW) to estimate C_{anth} change. Our results were nearly identical to those from the earlier 16 regression equations approach (Carter et al., 2017, 2019). Thus, we compared this method with other approaches using the same predictors (θ, S, AOU, Si, and N). We considered multiple (25) isopycnal intervals along

the sigma-0, sigma-2, and sigma-4 surface (Gruber 1998; Lee et al., 2003; Wanninkhof et al., 2010; Woosley et al., 2016), 14 isoneutral slabs (Gruber et al., 2019b), or no water mass division. In the meantime, we also used DIC (Friis et al., 2005; Wanninkhof et al., 2010), C* (Gruber et al., 1996, 2019a), and C^ (Carter et al., 2019) as the dependent variable of multiple linear regression.

In general, for both A16S (Figure A1) and A13.5 (Figure S9), all approaches lead to similar ΔC_{anth} patterns. The criteria used to identify water masses impacts the ΔC_{anth} estimations. The C*, C^ and DIC results of 14 isoneutral slabs (Figure A1(a), A1(b) and A1(c)) were similar to those based on our characteristic water masses (Figure A1(d), A1(e) and A1(f)). The 14 isoneutral slabs and characteristic water masses divide the ocean into several layers based on neutral density (γ). Overall, the estimations of ΔC_{anth} without separation of density layers (Figure A1(g)) showed lower variation in the upper ocean compared with other treatments and a more evenly distributed ΔC_{anth} from surface to deep depths. The ΔC_{anth} pattern using 25 isopycnal intervals (Figure A1(h)) generated similar results with the characteristic water masses (Figure A1(i)), but with more discontinuities. There may be too many separations for the 25 isopycnal intervals, resulting in insufficient data points in each density ranges. Thus, some of the established MLR equations may not be significant (leading to higher RMSE, see Additional supporting information: ASI_RMSE_and_residuals_for_Appendix.pptx). A higher ΔC_{anth} was located near 45-53°S in the surface layer and deeper penetrated with decreasing concentrations toward the interior down to 2000 m when 25 isopycnal intervals were applied (Figure A1(H)). Wanninkhof et al. (2010) and Woosley et al. (2016) also observed this deep penetration. As this deep penetration only appears when using 25 isopycnal intervals, we suspect that this is a method related artifact and suggest further study. The RMSE of using 14 slabs was close to that of using characteristic water masses division. The RMSE of not separating density layers was higher than that of the characteristic water masses (Additional supporting information: ASI_RMSE_and_residuals_for_Appendix.pptx). Therefore, it is reasonable for us to choose the 5 layers with six characteristic water masses as the basis of our approach.

Since dependent variable DIC, C* and C^ have been used to established multiple linear regression to estimated ΔC_{anth} in different studies (e.g., Wanninkhof et al., 2010; Clement and Gruber, 2018; Carter et al., 2019), we also compared the ΔC_{anth} with different dependent variables but using the same water masses separation (Figure A1(a), A1(b) and A1(c); or Figure A1(d), A1(e) and A1(f)). The change in DIC between two occupations (i.e., Figure 3a & 3b) is caused by not only the increase in C_{anth} (i.e., Figure 4 & 5) but also changes in the natural CO_2. The semi-conservative tracer C* is supposed to eliminate most of the natural CO_2 signal according to stoichiometric ratios. As Clement & Gruber (2018) mentioned, changes in C* are much closer to those of C_{anth}, resulting in separating C_{anth} from the background variability easier and statistically more robust. C^ is calculated similarly as C*, except no adjustments are applied for carbonate cycling or denitrification (Carter et al., 2019). ΔC_{anth} estimated by C* (Figure A1(a) & A1(d)) was lower in the intermediated waters (around 1000-2200m) in subantarctic region (~25-50°S) and in bottom waters between 42°S and 55°S than that estimated by C^ (Figure A1(b) & A1(e)) and DIC (Figure A1(c) & A1(f)). Although the order of RMSE along A16S from small to large was DIC < C^ < C*, their RMSE values were not significantly different (Additional supporting information: ASI_RMSE_and_residuals_for_Appendix.pptx). While our comparison here is not conclusive on which dependent variable is a better choice, we chose to use C* in order for our results to be comparable with the recent community effort (Gruber et al. 2019a).

ΔC_{anth} estimation is also sensitive to the choice of independent variables (Plancherel et al., 2013). It has been argued that ΔC_{anth} using the mean of several combinations of predictors for the

regressions decreases the errors and RMSE compared to using only one regression (Carter et al., 2017). When we applied the same 16 combinations of the five predictors for the regressions from Carter et al. (2019), however, not all these 16 regressions resulted in good RMSE and residuals. Regression 4, 8, 12 and 16 (those do not include AOU or N) had higher RMSE and residuals than the others (Additional supporting information: ASI_RMSE_and_residuals_for_Appendix.pptx). We felt it may be more reasonable that instead of using all 16 regressions, the mean of 10-12 best regressions should be used as the final ΔC_{anth}. Thus, similar to Clement & Gruber (2018) and Gruber et al. (2019), we chose the 11 best fits from over 100 possible combinations with possible predictors according to not only small RMSE but also multiple and partial correlation coefficients (see details of this method in the supplementary Text S2). The mean RMSE of our 11 best combinations was better than that of 16 combinations.

Figure A1. Comparison of anthropogenic CO_2 change along the A16S transect for the time interval between 2013 and 1989. Nine cases are considered: (a) C* is used as dependent variable, water mass is divided by 14 isoneutral slabs and 16 regressions are considered. (b) & (c) are similar to (a), but use C^ and DIC as dependent variable, separately. (d) C* is used as dependent variable, water mass is divided by 5 layers with six characteristic water masses and 16 regressions are considered. Similarly, (e) & (f) use C^ and DIC as dependent variable. (g) uses DIC as dependent variable, but does not divide water mass and does not adopt 16 regressions. (h) uses DIC as dependent variable, and uses 25 isopycnal intervals, but does not adopt 16 regressions. (i) uses DIC as dependent variable, and uses 5 layers with six characteristic water masses, but does not adopt 16 regressions.

We also examined whether different variables used in the MLR have strong inherent dependencies. In general, all of the 11 best combinations contain AOU, the difference lies in the choice of temperature or density, the selection of nutrients and whether to use TA as a predictor. Temperature (or potential temperature) showed a good correlation with density (or potential density) ($r>0.9$), and thus it is better not to use them as predictors at the same time. The correlation coefficient between nitrate and phosphate was also very high ($r>0.99$), implying that only one nutrient should be chosen (Supplementary materials).

For transects A13.5 (Figure S10), A16S between 2013 and 2005 (Figure S12), and A10 between 2011 and 2003 (Figure S15), the patterns of ΔC_{anth} from regression 1-11 matched very closely with each other. But for transect A16S between 2013 and 1989 (Figure S11), as well as between 2005 and 1989 (Figure S13), the patterns of ΔC_{anth} did not always agree, especially when using TA as a predictor (Reg #8 ~ Reg #11). Woosley et al. (2016) preferred to use the eMLR with TA than Si, because, by this approach, the resulting pattern of ΔC_{anth} matches very closely to the transient tracer SF_6 measured on A16S (2013). In our study, ΔC_{anth} of A16S (2005-2013) estimated with TA (Figure S12, Reg #8 ~ Reg #11) or Si (Figure S12, Reg #1 ~ Reg #2 and Reg #5 ~ Reg #7) were similar and also matched closely with tracer CFC-12 (Figure 2j). However, estimated ΔC_{anth} along A16S (1989-2013 or 1989-2005) were different whether TA or Si is used (Figure S11 and Figure S13). This could be caused by the low quality of TA in the 1989 cruise as suggested by Woosley et al. (2016). More repeat observation data or models are needed to further assess and resolve this discrepancy.

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References

Abrahamsen, E. P., Meijers, A. J., Polzin, K. L., Garabato, A. C. N., King, B. A., Firing, Y. L., ... & Meredith, M. P. (2019). Stabilization of dense Antarctic water supply to the Atlantic Ocean overturning circulation. Nature Climate Change, 9(10), 742-746. https://doi.org/10.1038/s41558-019-0561-2

Anderson, L. A., & Sarmiento, J. L. (1994). Redfield ratios of remineralization determined by nutrient data analysis. Global biogeochemical cycles, 8(1), 65-80. https://doi.org/10.1029/93GB03318

Arhan, M., Mercier, H., & Park, Y. H. (2003). On the deep water circulation of the eastern South Atlantic Ocean. Deep Sea Research Part I: Oceanographic Research Papers, 50(7), 889-916. https://doi.10.1016/s0967-0637(03)00072-4

Brewer, P. G. (1978). Direct observation of the oceanic CO2 increase. Geophysical Research Letters, 5(12), 997-1000. https://doi.org/10.1029/GL005i012p00997

Bryden, H. L., King, B. A., & McCarthy, G. D. (2011). South Atlantic overturning circulation at 24 S. Journal of Marine Research, 69(1), 38-55. https://doi.org/10.1357/002224011798147633

Caldeira, K., & Duffy, P. B. (2000). The role of the Southern Ocean in uptake and storage of anthropogenic carbon dioxide. Science, 287(5453), 620-622. https://doi.org/10.1126/science.287.5453.620

Carter, B. R., Feely, R. A., Mecking, S., Cross, J. N., Macdonald, A. M., Siedlecki, S. A., ... & Rodgers, K. B. (2017). Two decades of Pacific anthropogenic carbon storage and ocean acidification along Global Ocean Ship-based Hydrographic Investigations Program sections P16 and P02. Global Biogeochemical Cycles, 31(2), 306-327. https://doi.org/10.1002/2016GB005485

Carter, B. R., Feely, R. A., Wanninkhof, R., Kouketsu, S., Sonnerup, R. E., Pardo, P. C., ... & Bullister, J. L. (2019). Pacific anthropogenic carbon between 1991 and 2017. Global Biogeochemical Cycles, 33(5), 597-617. https://doi.org/10.1029/2018GB006154

Chen, G. T., & Millero, F. J. (1979). Gradual increase of oceanic CO2. Nature, 277(5693), 205-206. https://doi.org/10.1038/277205a0

Clement, D., & Gruber, N. (2018). The eMLR (C*) method to determine decadal changes in the global ocean storage of anthropogenic CO2. Global Biogeochemical Cycles, 32(4), 654-679. https://doi.org/10.1002/2017GB005819

DeVries, T., Holzer, M., & Primeau, F. (2017). Recent increase in oceanic carbon uptake driven by weaker upper-ocean overturning. Nature, 542(7640), 215-218. https://doi.org/10.1038/nature21068

Dickson, A. G., & Millero, F. J. (1987). A comparison of the equilibrium constants for the dissociation of carbonic acid in seawater media. Deep Sea Research Part A. Oceanographic Research Papers, 34(10), 1733-1743. https://doi.org/10.1016/0198-0149(87)90021-5

Doney, S. C., & Hecht, M. W. (2002). Antarctic bottom water formation and deep-water chlorofluorocarbon distributions in a global ocean climate model. Journal of Physical

Oceanography, 32(6), 1642-1666. https://doi.org/10.1175/1520-0485(2002)032<1642:ABWFAD>2.0.CO;2

Doney, S. C., Fabry, V. J., Feely, R. A., & Kleypas, J. A. (2009). Ocean acidification: the other CO2 problem. Annual review of marine science, 1, 169-192. https://doi.org/10.1146/annurev.marine.010908.163834

Doney, S. C. (2010). The growing human footprint on coastal and open-ocean biogeochemistry. science, 328(5985), 1512-1516. https://doi.org/10.1126/science.1185198

Dong, S., Garzoli, S., Baringer, M., Meinen, C., & Goni, G. (2009). Interannual variations in the Atlantic meridional overturning circulation and its relationship with the net northward heat transport in the South Atlantic. Geophysical Research Letters, 36(20). https://doi.org/10.1029/2009GL039356

Evans, G. R., McDonagh, E. L., King, B. A., Bryden, H. L., Bakker, D. C. E., Brown, P. J., ... & Van Heuven, S. M. A. C. (2017). South Atlantic interbasin exchanges of mass, heat, salt and anthropogenic carbon. Progress in Oceanography, 151, 62-82. https://doi.org/10.1016/j.pocean.2016.11.005

Feely, R. A., Sabine, C. L., Lee, K., Berelson, W., Kleypas, J., Fabry, V. J., & Millero, F. J. (2004). Impact of anthropogenic CO_2 on the $CaCO_3$ system in the oceans. Science, 305(5682), 362-366. https://doi.org/10.1126/science.1097329

Feucher, C., Maze, G., & Mercier, H. (2019). Subtropical mode water and permanent pycnocline properties in the world ocean. Journal of Geophysical Research: Oceans, 124(2), 1139-1154. https://doi.org/10.1029/2018JC014526

Fine, R. A. (2011). Observations of CFCs and SF6 as ocean tracers. Annual Review of Marine Science, 3, 173-195. https://doi.org/10.1146/annurev.marine.010908.163933

Friedlingstein, P., Jones, M. W., O'sullivan, M., Andrew, R. M., Hauck, J., Peters, G. P., ... & Zaehle, S. (2019). Global carbon budget 2019. Earth System Science Data, 11(4), 1783-1838. https://doi.org/10.5194/essd-11-1783-2019

Friedlingstein, P., O'sullivan, M., Jones, M. W., Andrew, R. M., Hauck, J., Olsen, A., ... & Zaehle, S. (2020). Global carbon budget 2020. Earth System Science Data, 12(4), 3269-3340. https://doi.org/10.5194/essd-12-3269-2020

Friis, K., Körtzinger, A., Pätsch, J., & Wallace, D. W. (2005). On the temporal increase of anthropogenic CO2 in the subpolar North Atlantic. Deep Sea Research Part I: Oceanographic Research Papers, 52(5), 681-698. https://doi.org/10.1016/j.dsr.2004.11.017

Gordon, A. L., & Greengrove, C. L. (1986). Geostrophic circulation of the Brazil-Falkland confluence. Deep Sea Research Part A. Oceanographic Research Papers, 33(5), 573-585. https://doi.org/10.1016/0198-0149(86)90054-3

Gruber, N., Sarmiento, J. L., & Stocker, T. F. (1996). An improved method for detecting anthropogenic CO2 in the oceans. Global Biogeochemical Cycles, 10(4), 809-837. https://doi.org/10.1029/96GB01608

Gruber, N. (1998). Anthropogenic CO_2 in the Atlantic Ocean. Global Biogeochemical Cycles, 12(1), 165-191. https://doi.org/10.1029/97GB03658

Gruber, N. I. C. O. L. A. S., & Sarmiento, J. L. (2002). Large-scale biogeochemical-physical interactions in elemental cycles. The sea, 12, 337-399.

Gruber, N., Gloor, M., Mikaloff Fletcher, S. E., Doney, S. C., Dutkiewicz, S., Follows, M. J., ... & Takahashi, T. (2009). Oceanic sources, sinks, and transport of atmospheric CO2. Global biogeochemical cycles, 23(1). https://doi.org/10.1029/2008GB003349

Gruber, N., Clement, D., Carter, B. R., Feely, R. A., Van Heuven, S., Hoppema, M., ... & Wanninkhof, R. (2019a). The oceanic sink for anthropogenic CO2 from 1994 to 2007. Science, 363(6432), 1193-1199. https://doi.org/10.1126/science.aau5153

Gruber, N., Landschützer, P., & Lovenduski, N. S. (2019b). The variable Southern Ocean carbon sink. Annual review of marine science, 11, 159-186. https://doi.org/10.1146/annurev-marine-121916-063407

Goodkin, N. F., Levine, N. M., Doney, S. C., & Wanninkhof, R. (2011). Impacts of temporal CO2 and climate trends on the detection of ocean anthropogenic CO2 accumulation. Global Biogeochemical Cycles, 25(3). https://doi.org/10.1029/2010GB004009

Herraiz-Borreguero, L., & Rintoul, S. R. (2011). Subantarctic mode water: distribution and circulation. Ocean Dynamics, 61(1), 103-126. https://doi.org/10.1007/s10236-010-0352-9

Heywood, K. J., & King, B. A. (2002). Water masses and baroclinic transports in the South Atlantic and Southern oceans. Journal of Marine Research, 60(5), 639-676. https://doi.org/10.1357/002224002762688687

Hogg, N. G., & Thurnherr, A. M. (2005). A zonal pathway for NADW in the South Atlantic. Journal of oceanography, 61(3), 493-507. https://doi.org/10.1007/s10872-005-0058-7

Holte, J., Talley, L. D., Gilson, J., & Roemmich, D. (2017). An Argo mixed layer climatology and database. Geophysical Research Letters, 44(11), 5618-5626. https://doi.org/10.1002/2017GL073426

Iudicone, D., Rodgers, K. B., Stendardo, I., Aumont, O., Madec, G., Bopp, L., ... & Ribera d'Alcala, M. (2011). Water masses as a unifying framework for understanding the Southern Ocean Carbon Cycle. Biogeosciences, 8(5), 1031-1052. https://doi.org/10.5194/bg-8-1031-2011

Iudicone, D., Rodgers, K. B., Plancherel, Y., Aumont, O., Ito, T., Key, R. M., ... & Ishii, M. (2016). The formation of the ocean's anthropogenic carbon reservoir. Scientific reports, 6(1), 1-16. https://doi.org/10.1038/srep35473

Keeling, C. D. (1986). Atmospheric CO2 concentrations-Mauna Loa Observatory, Hawaii 1958-1986 (No. 2798). Oak Ridge National Laboratory.

Khatiwala, S., Primeau, F., & Hall, T. (2009). Reconstruction of the history of anthropogenic CO 2 concentrations in the ocean. Nature, 462(7271), 346-349. https://doi.org/10.1038/nature08526

Khatiwala, S., Tanhua, T., Mikaloff Fletcher, S., Gerber, M., Doney, S. C., Graven, H. D., ... & Sabine, C. L. (2013). Global ocean storage of anthropogenic carbon. Biogeosciences, 10(4), 2169-2191. https://doi.org/10.5194/bg-10-2169-2013

Kleypas, J. A., Buddemeier, R. W., Archer, D., Gattuso, J. P., Langdon, C., & Opdyke, B. N. (1999). Geochemical consequences of increased atmospheric carbon dioxide on coral reefs. science, 284(5411), 118-120. https://doi.org/10.1126/science.284.5411.118

Kouketsu, S., & Murata, A. M. (2014). Detecting decadal scale increases in anthropogenic CO2 in the ocean. Geophysical Research Letters, 41(13), 4594-4600. https://doi.org/10.1002/2014GL060516

Kraus, E. B., & Businger, J. A. (1994). Atmosphere-ocean interaction. Oxford University Press.

Landschützer, Peter; Gruber, Nicolas; Bakker, Dorothee C. E. (2020). An observation-based global monthly gridded sea surface pCO2 product from 1982 onward and its monthly climatology (NCEI Accession 0160558). Version 5.5. NOAA National Centers for Environmental Information. Dataset. https://doi.org/10.7289/V5Z899N6. [2020-07-10]

Landschützer, P., Gruber, N., & Bakker, D. C. (2016). Decadal variations and trends of the global ocean carbon sink. Global Biogeochemical Cycles, 30(10), 1396-1417. https://doi.org/10.1002/2015GB005359

Landschützer, P., Gruber, N., Haumann, F. A., Rödenbeck, C., Bakker, D. C., van Heuven, S., ... & Wanninkhof, R. (2015). The reinvigoration of the Southern Ocean carbon sink. Science, 349(6253), 1221-1224. https://doi.org/10.1126/science.aab2620

Le Quéré, C., Rödenbeck, C., Buitenhuis, E. T., Conway, T. J., Langenfelds, R., Gomez, A., ... & Heimann, M. (2007). Saturation of the Southern Ocean CO2 sink due to recent climate change. science, 316(5832), 1735-1738. https://doi.org/10.1126/science.1136188

Le Quéré, C., Andrew, R. M., Canadell, J. G., Sitch, S., Korsbakken, J. I., Peters, G. P., ... & Zaehle, S. (2016). Global carbon budget 2016. Earth System Science Data (Online), 8(2). https://doi.org/10.5194/essd-8-605-2016

Lee, K., Choi, S. D., Park, G. H., Wanninkhof, R., Peng, T. H., Key, R. M., ... & Kozyr, A. (2003). An updated anthropogenic CO_2 inventory in the Atlantic Ocean. Global Biogeochemical Cycles, 17(4). https://doi.org/10.1029/2003GB002067

Levine, N. M., Doney, S. C., Wanninkhof, R., Lindsay, K., & Fung, I. Y. (2008). Impact of ocean carbon system variability on the detection of temporal increases in anthropogenic CO2. Journal of Geophysical Research: Oceans, 113(C3). https://doi.org/10.1029/2007JC004153

Marshall, J., & Speer, K. (2012). Closure of the meridional overturning circulation through Southern Ocean upwelling. Nature Geoscience, 5(3), 171-180. https://doi.org/10.1038/ngeo1391

McCartney, M. S., 1977. Subantarctic Mode Water. In: A Voyage of Discovery: George Deacon 70th Anniversary Volume, M. V. Angel, editor, Supplement to Deep-Sea Research, Pergamon Press, Oxford, pp. 103-119.

McNeil, B. I., Tilbrook, B., & Matear, R. J. (2001). Accumulation and uptake of anthropogenic CO_2 in the Southern Ocean, south of Australia between 1968 and 1996. Journal of Geophysical Research: Oceans, 106(C12), 31431-31445. https://doi.org/10.1029/2000JC000331

Mémery, L., Arhan, M., Álvarez-Salgado, X. A., Messias, M. J., Mercier, H., Castro, C. G., & Rios, A. F. (2000). The water masses along the western boundary of the south and equatorial Atlantic. Progress in Oceanography, 47(1), 69-98. https://doi.org/10.1016/S0079-6611(00)00032-X

Messias, M. J., & Laurent Memery, L. O. D. Y. C. (1998). South Atlantic chlorofluoromethane distributions along the WHP lines A17, A13 and A14. WOCE, 21.

Mercier, H., Arhan, M., & Lutjeharms, J. R. (2003). Upper-layer circulation in the eastern Equatorial and South Atlantic Ocean in January–March 1995. Deep Sea Research Part I: Oceanographic Research Papers, 50(7), 863-887. https://doi.org/10.1016/S0967-0637(03)00071-2

Millero, F. J. (2007). The marine inorganic carbon cycle. Chemical reviews, 107(2), 308-341. https://doi.org/10.1021/cr0503557

Murata, A., Kumamoto, Y., Sasaki, K. I., Watanabe, S., & Fukasawa, M. (2008). Decadal increases of anthropogenic CO_2 in the subtropical South Atlantic Ocean along 30 S. Journal of Geophysical Research: Oceans, 113(C6). https://doi.org/10.1029/2007JC004424

Olsen, A., Lange, N., Key, R. M., Tanhua, T., Álvarez, M., Becker, S., Bittig, H. C., Carter, B. R., Cotrim da Cunha, L., Feely, R. A., van Heuven, S., Hoppema, M., Ishii, M., Jeansson, E., Jones, S. D., Jutterström, S., Karlsen, M. K., Kozyr, A., Lauvset, S. K., Lo Monaco, C., Murata, A., Pérez, F. F., Pfeil, B., Schirnick, C., Steinfeldt, R., Suzuki, T., Telszewski, M., Tilbrook, B.,

Velo, A. and Wanninkhof, R.(2019). GLODAPv2.2019 – an update of GLODAPv2. Earth System Science Data, 11(3), 1437-1461. doi:10.5194/essd-11-1437-2019.

Orr, J. C., Fabry, V. J., Aumont, O., Bopp, L., Doney, S. C., Feely, R. A., ... & Yool, A. (2005). Anthropogenic ocean acidification over the twenty-first century and its impact on calcifying organisms. Nature, 437(7059), 681-686. https://doi.org/10.1038/nature04095

Orsi, A. H., Whitworth III, T., & Nowlin Jr, W. D. (1995). On the meridional extent and fronts of the Antarctic Circumpolar Current. Deep Sea Research Part I: Oceanographic Research Papers, 42(5), 641-673. https://doi.org/10.1016/0967-0637(95)00021-W

Orsi, A. H., Johnson, G. C., & Bullister, J. L. (1999). Circulation, mixing, and production of Antarctic Bottom Water. Progress in Oceanography, 43(1), 55-109. https://doi.org/10.1016/S0079-6611(99)00004-X

Pardo, P. C., Pérez, F. F., Khatiwala, S., & Ríos, A. F. (2014). Anthropogenic CO2 estimates in the Southern Ocean: Storage partitioning in the different water masses. Progress in Oceanography, 120, 230-242. https://doi.org/10.1016/j.pocean.2013.09.005

Peng, T. H., & Wanninkhof, R. (2010). Increase in anthropogenic CO2 in the Atlantic Ocean in the last two decades. Deep Sea Research Part I: Oceanographic Research Papers, 57(6), 755-770. https://doi.org/10.1016/j.dsr.2010.03.008

Pérez, F. F., Mercier, H., Vázquez-Rodríguez, M., Lherminier, P., Velo, A., Pardo, P. C., ... & Ríos, A. F. (2013). Atlantic Ocean CO 2 uptake reduced by weakening of the meridional overturning circulation. Nature Geoscience, 6(2), 146-152. https://doi.org/10.1038/ngeo1680

Pérez, F. F., Fontela, M., García-Ibáñez, M. I., Mercier, H., Velo, A., Lherminier, P., ... & Padin, X. A. (2018). Meridional overturning circulation conveys fast acidification to the deep Atlantic Ocean. Nature, 554(7693), 515-518. https://doi.org/10.1038/nature25493

Peterson, R. G., & Stramma, L. (1991). Upper-level circulation in the South Atlantic Ocean. Progress in oceanography, 26(1), 1-73. https://doi.org/10.1016/0079-6611(91)90006-8

Plancherel, Y., Rodgers, K. B., Key, R. M., Jacobson, A. R., & Sarmiento, J. L. (2013). Role of regression model selection and station distribution on the estimation of oceanic anthropogenic carbon change by eMLR. Biogeosciences, 10(7), 4801-4831. https://doi.org/10.5194/bg-10-4801-2013

Quay, P., Sonnerup, R., Stutsman, J., Maurer, J., Körtzinger, A., Padin, X. A., & Robinson, C. (2007). Anthropogenic CO2 accumulation rates in the North Atlantic Ocean from changes in the 13C/12C of dissolved inorganic carbon. Global biogeochemical cycles, 21(1). https://doi.org/10.1029/2006GB002761

Quay, P., Sonnerup, R., Munro, D., & Sweeney, C. (2017). Anthropogenic CO2 accumulation and uptake rates in the Pacific Ocean based on changes in the 13C/12C of dissolved inorganic carbon. Global Biogeochemical Cycles, 31(1), 59-80. https://doi.org/10.1002/2016GB005460

Rhein, M., Stramma, L., & Krahmann, G. (1998). The spreading of Antarctic bottom water in the tropical Atlantic. Deep Sea Research Part I: Oceanographic Research Papers, 45(4-5), 507-527. https://doi.org/10.1016/S0967-0637(97)00030-7

Ríos, A. F., Álvarez-Salgado, X. A., Pérez, F. F., Bingler, L. S., Arístegui, J., & Mémery, L. (2003). Carbon dioxide along WOCE line A14: Water masses characterization and anthropogenic entry. Journal of Geophysical Research: Oceans, 108(C4). https://doi.org/10.1029/2000JC000366

Ríos, A. F., Vázquez-Rodríguez, M., Padín, X. A., & Pérez, F. F. (2010). Anthropogenic carbon dioxide in the South Atlantic western basin. Journal of Marine Systems, 83(1-2), 38-44. https://doi.org/10.1016/j.jmarsys.2010.06.010

Ríos, A. F., Velo, A., Pardo, P. C., Hoppema, M., & Pérez, F. F. (2012). An update of anthropogenic CO_2 storage rates in the western South Atlantic basin and the role of Antarctic Bottom Water. Journal of Marine Systems, 94, 197-203. https://doi.org/10.1016/j.jmarsys.2011.11.023

Sabine, C. L., Feely, R. A., Gruber, N., Key, R. M., Lee, K., Bullister, J. L., ... & Millero, F. J. (2004). The oceanic sink for anthropogenic CO_2. science, 305(5682), 367-371. https://doi.org/10.1126/science.1097403

Sabine, C. L., Feely, R. A., Millero, F. J., Dickson, A. G., Langdon, C., Mecking, S., & Greeley, D. (2008). Decadal changes in Pacific carbon. Journal of Geophysical Research: Oceans, 113(C7). https://doi.org/10.1029/2007JC004577

Sabine, C. L., & Tanhua, T. (2010). Estimation of anthropogenic CO_2 inventories in the ocean. Annual review of marine science, 2, 175-198. https://doi.org/10.1146/annurev-marine-120308-080947

Santoso, A., England, M. H., & Hirst, A. C. (2006). Circumpolar deep water circulation and variability in a coupled climate model. Journal of physical oceanography, 36(8), 1523-1552. https://doi.org/10.1175/JPO2930.1

Sallée, J. B., Speer, K., Rintoul, S., & Wijffels, S. (2010). Southern Ocean thermocline ventilation. Journal of Physical Oceanography, 40(3), 509-529. https://doi.org/10.1175/2009JPO4291.1

Sallée, J. B., Matear, R. J., Rintoul, S. R., & Lenton, A. (2012). Localized subduction of anthropogenic carbon dioxide in the Southern Hemisphere oceans. Nature Geoscience, 5(8), 579-584. https://doi.org/10.1038/ngeo1523

Salt, L. A., van Heuven, S. M. A. C., Claus, M. E., Jones, E. M., & de Baar, H. J. W. (2014). Rapid acidification of mode and intermediate waters in the southwest Atlantic Ocean. Biogeosciences Discussions, 11, 6755-6792. https://doi.org/10.5194/bg-12-1387-2015

Shillington, F. A. (1998). The Benguela upwelling system off southwestern Africa. The sea, 11, 583-604.

Takahashi, T., Sutherland, S. C., Wanninkhof, R., Sweeney, C., Feely, R. A., Chipman, D. W., ... & De Baar, H. J. (2009). Climatological mean and decadal change in surface ocean pCO2, and net sea–air CO2 flux over the global oceans. Deep Sea Research Part II: Topical Studies in Oceanography, 56(8-10), 554-577. https://doi.org/10.1016/j.dsr2.2008.12.009

Tanhua, T., & Keeling, R. F. (2012). Changes in column inventories of carbon and oxygen in the Atlantic Ocean. Biogeosciences (BG), 9, 4819-4833.

Tanhua, T., Hoppema, M., Jones, E. M., Stöven, T., Hauck, J., Dávila, M. G., ... & Strass, V. H. (2017). Temporal changes in ventilation and the carbonate system in the Atlantic sector of the Southern Ocean. Deep Sea Research Part II: Topical Studies in Oceanography, 138, 26-38. https://doi.org/10.5194/bg-9-4819-2012

Talley, L. D. (2011). Descriptive physical oceanography: an introduction. Academic press.

Touratier, F., & Goyet, C. (2004). Applying the new TrOCA approach to assess the distribution of anthropogenic CO2 in the Atlantic Ocean. Journal of Marine Systems, 46(1-4), 181-197. https://doi.org/10.1016/j.jmarsys.2003.11.020

Tsuchiya, M., Talley, L. D., & McCartney, M. S. (1994). Water-mass distributions in the western South Atlantic; A section from South Georgia Island (54S) northward across the equator. Journal of Marine Research, 52(1), 55-81. https://doi.org/10.1357/0022240943076759

Vázquez-Rodríguez, M., Touratier, F., Monaco, C. L., Waugh, D. W., Padin, X. A., Bellerby, R. G. J., ... & Pérez, F. F. (2009). Anthropogenic carbon distributions in the Atlantic Ocean:

1176 data-based estimates from the Arctic to the Antarctic, Biogeosciences, 6, 439–451.
1177 https://doi.org/10.5194/bg-6-439-2009
1178 Wacongne, S., & Piton, B. (1992). The near-surface circulation in the northeastern corner of
1179 the South Atlantic Ocean. Deep Sea Research Part A. Oceanographic Research Papers, 39(7-8),
1180 1273-1298. https://doi.org/10.1016/0198-0149(92)90069-6
1181 Wanninkhof, R., Doney, S. C., Peng, T. H., Bullister, J. L., Lee, K., & Feely, R. A. (1999).
1182 Comparison of methods to determine the anthropogenic CO2 invasion into the Atlantic Ocean.
1183 Tellus B: Chemical and Physical Meteorology, 51(2), 511-530.
1184 https://doi.org/10.3402/tellusb.v51i2.16335
1185 Wanninkhof, R., Doney, S. C., Bullister, J. L., Levine, N. M., Warner, M., & Gruber, N.
1186 (2010). Detecting anthropogenic CO2 changes in the interior Atlantic Ocean between 1989 and
1187 2005. Journal of Geophysical Research: Oceans, 115(C11).
1188 https://doi.org/10.1029/2010JC006251
1189 Wallace, D. (1995). Monitoring global ocean carbon inventories, 54 pp. Ocean Obs. Syst.
1190 Dev. Panel, Texas A&M Univ., College Station, Tex.
1191 Williams, N. L., Feely, R. A., Sabine, C. L., Dickson, A. G., Swift, J. H., Talley, L. D., &
1192 Russell, J. L. (2015). Quantifying anthropogenic carbon inventory changes in the Pacific sector
1193 of the Southern Ocean. Marine Chemistry, 174, 147-160.
1194 https://doi.org/10.1016/j.marchem.2015.06.015
1195 Woosley, R. J., Millero, F. J., & Wanninkhof, R. (2016). Rapid anthropogenic changes in
1196 CO2 and pH in the Atlantic Ocean: 2003–2014. Global Biogeochemical Cycles, 30(1), 70-90.
1197 https://doi.org/10.1002/2015GB005248