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Key Points:

- Satellite and in situ observations with Lagrangian tracking were used to estimate the cumulative CO₂ flux of long lived mesoscale eddies
- Drivers of the $p\text{CO}_{2(\text{sw})}$ variability significantly changed over the anticyclonic eddies lifetime but did not in cyclonic eddies
- Both anticyclonic and cyclonic eddies enhance the CO₂ sink into the South Atlantic Ocean

Supporting Information:

Supporting Information may be found in the online version of this article.

Correspondence to:

D. J. Ford,
d.ford@exeter.ac.uk

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Author Contributions:

Conceptualization: Daniel J. Ford, Gavin H. Tilstone, Jamie D. Shutler, Vassilis Kitidis, Katy L. Sheen, Giorgio Dall'Omo

Data curation: Daniel J. Ford, Iole B. M. Orselli

Formal analysis: Daniel J. Ford

Funding acquisition: Gavin H. Tilstone, Jamie D. Shutler, Vassilis Kitidis

Investigation: Daniel J. Ford, Iole B. M. Orselli

Methodology: Daniel J. Ford, Gavin H. Tilstone, Jamie D. Shutler, Vassilis Kitidis, Katy L. Sheen

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Mesoscale Eddies Enhance the Air-Sea CO₂ Sink in the South Atlantic Ocean

Daniel J. Ford^{1,2,3} , Gavin H. Tilstone¹ , Jamie D. Shutler² , Vassilis Kitidis¹ , Katy L. Sheen², Giorgio Dall'Omo^{1,4} , and Iole B. M. Orselli⁵ 

¹Plymouth Marine Laboratory, Plymouth, UK, ²College of Life and Environmental Sciences, University of Exeter, Penryn, UK, ³Now at Faculty of Environment, Science and Economy, University of Exeter, Penryn, UK, ⁴Now at Istituto Nazionale di Oceanografia e di Geofisica Sperimentale, Trieste, Italy, ⁵Laboratório de Estudos dos Oceanos e Clima, Instituto de Oceanografia, Universidade Federal do Rio Grande (FURG), Rio Grande, Brazil

Abstract Mesoscale eddies are abundant in the global oceans and known to affect oceanic and atmospheric conditions. Understanding their cumulative impact on the air-sea carbon dioxide (CO₂) flux may have significant implications for the ocean carbon sink. Observations and Lagrangian tracking were used to estimate the air-sea CO₂ flux of 67 long lived (>1 year) mesoscale eddies in the South Atlantic Ocean over a 16 year period. Both anticyclonic eddies originating from the Agulhas retroflection and cyclonic eddies originating from the Benguela upwelling act as net CO₂ sinks over their lifetimes. Anticyclonic eddies displayed an exponential decrease in the net CO₂ sink, whereas cyclonic eddies showed a linear increase. Combined, these eddies significantly enhanced the CO₂ sink into the South Atlantic Ocean by $0.08 \pm 0.04\%$. The studied eddies constitute a fraction of global eddies, and eddy activity is increasing; therefore, explicitly resolving eddies appears critical when assessing the ocean carbon sink.

Plain Language Summary Ocean mesoscale eddies can form when part of a main current becomes separated or through internal ocean instabilities which form circular rotating currents that propagate across the oceans. These eddies last from weeks to years and can modify the ocean properties of the water captured within them, which in turn affects the net exchange of carbon between this water and the atmosphere. Little is known about how these eddies modify the absorption of carbon across the global ocean, collectively referred to as the ocean carbon sink, despite them being ubiquitous features of the global oceans. Using in situ and satellite-based observations, we show that eddies in the South Atlantic Ocean enhance the absorption of carbon from the atmosphere, thus modifying the ocean to be a stronger net sink of carbon. These results are important as they quantify how much eddies contribute to the absorption of carbon from the atmosphere to the ocean, and highlight the need to include eddies when assessing ocean carbon budgets.

1. Introduction

Mesoscale eddies, characterized by radii on the order of 100 kilometers (km) and lifetimes of weeks to years, are ubiquitous in the global oceans (Chelton et al., 2011; Pegliasco et al., 2022). Eddies modify the physical (Laxenaire et al., 2019; Nencioli et al., 2018), biological (Carvalho et al., 2019; Dufois et al., 2016; Lehahn et al., 2011; Roughan et al., 2017), and chemical (Arhan et al., 2011; Chen et al., 2007; Orselli, Goyet, et al., 2019; Orselli, Kerr, et al., 2019) characteristics of the ocean compared to the surrounding waters and can be advected far away from their origin. Alteration of the ocean surface conditions can modulate the air-sea exchange of CO₂ through changes in the partial pressure of CO₂ ($p\text{CO}_{2(\text{sw})}$) (Chen et al., 2007; Jones et al., 2017; Orselli, Kerr, et al., 2019; Song et al., 2016), solubility of CO₂, and the overlying atmospheric conditions (Frenger et al., 2013; Pezzi et al., 2021; Souza et al., 2021). Despite their abundance, few studies have investigated the role of eddies in the air-sea exchange of CO₂ (Chen et al., 2007; Jones et al., 2017; Pezzi et al., 2021) and estimated their cumulative impact on the oceanic CO₂ sink (Orselli, Kerr, et al., 2019).

Anticyclonic eddies generally display high-pressure centers, displace isopycnals downwards inducing downwelling, and have higher sea surface temperatures (SST) than the surrounding environment (McGillicuddy, 2016). The CO₂ solubility in seawater decreases with increasing temperature (Weiss, 1974), and biological activity would hypothetically decrease due to lower nutrient inputs into the surface layer caused by downwelling (Angel-Benavidez et al., 2016; Gaube et al., 2014; Liu et al., 2018). Therefore, these anticyclonic features could

Project Administration: Daniel J. Ford, Gavin H. Tilstone, Jamie D. Shutler, Vassilis Kitidis, Katy L. Sheen
Software: Daniel J. Ford
Supervision: Gavin H. Tilstone, Jamie D. Shutler, Vassilis Kitidis, Katy L. Sheen
Validation: Daniel J. Ford
Visualization: Daniel J. Ford
Writing – original draft: Daniel J. Ford
Writing – review & editing: Daniel J. Ford, Gavin H. Tilstone, Jamie D. Shutler, Vassilis Kitidis, Katy L. Sheen, Giorgio Dall’Omo, Iole B. M. Orselli

increase $p\text{CO}_{2(\text{sw})}$ and act as weak CO_2 sinks or even sources of CO_2 to the atmosphere. Cyclonic eddies follow the opposite convention with low-pressure centers, lower SST (Chen et al., 2007), elevated isopycnals inducing upwelling, enhanced biological activity due to upwelled nutrients (Angel-Benavidez et al., 2016) and therefore potentially decreasing $p\text{CO}_{2(\text{sw})}$, and enhancing the CO_2 sink.

Mesoscale eddies are, however, intricate structures, and the way they modify the air-sea CO_2 fluxes is more complex. Jones et al. (2017) identified that both anticyclonic and cyclonic eddies were hotspots for CO_2 draw-down in the Southern Ocean. Orselli, Kerr, et al. (2019) showed that anticyclonic (Agulhas) eddies are a stronger CO_2 sink than the surrounding water in the South Atlantic. By contrast, Pezzi et al. (2021) identified an anticyclonic eddy as a CO_2 source and Chen et al. (2007) reported that a single cyclonic eddy in the North Pacific weakened the CO_2 sink by $\sim 17\%$. Song et al. (2016) showed that the way in which eddies modify the air-sea CO_2 flux can change seasonally in the Southern Ocean: anticyclonic eddies were stronger CO_2 sources in winter and stronger CO_2 sinks in summer, and the opposite was found for cyclonic eddies. The ability of mesoscale eddies to modify the CO_2 flux as they age (Orselli, Kerr, et al., 2019), may also vary seasonally.

The South Atlantic Ocean contributes $\sim 10\%$ to the global ocean CO_2 sink (e.g., Landschützer et al., 2014). The basin has some of the largest long-lived (>1 year) anticyclonic eddies globally, originating from the Agulhas retroflection (Lutjeharms, 2006), and can propagate to the Brazilian Coast (Guerra et al., 2018). In conjunction, cyclonic eddies from the Benguela upwelling system propagate across the basin (Chelton et al., 2011; Pegliasco et al., 2022; Rubio et al., 2009). The effect of eddies on the air-sea CO_2 flux, differences between anticyclonic and cyclonic, and their role in the global ocean CO_2 sink requires further investigation, especially since eddy kinetic energy has been increasing globally (Martínez-Moreno et al., 2021).

The objective of this study is to estimate the air-sea CO_2 flux of long-lived mesoscale eddies in the South Atlantic Ocean using satellite and in situ observations. A total of 67 eddies, 36 Agulhas anticyclonic and 31 Benguela cyclonic, were tracked using satellite observations (2002–2018) and Lagrangian cumulative air-sea CO_2 fluxes were estimated in order to assess their role in the South Atlantic CO_2 sink. To help understand which processes are controlling the change in flux over the lifetime of each eddy, the $p\text{CO}_{2(\text{sw})}$ timeseries for each eddy was decomposed into the thermal and non-thermal drivers.

2. Data and Methods

2.1. Sea Surface Temperature, Salinity, Biological and Wind Speed Data

Daily 4 km chlorophyll *a* (chl *a*) composites were calculated from Moderate Resolution Imaging Spectroradiometer on Aqua (MODIS-A) Level 1 granules, downloaded from the National Aeronautics Space Administration (NASA) Ocean Color website (<https://oceancolor.gsfc.nasa.gov/>; accessed 10 December 2020), using SeaDAS v7.5, and applying the standard OC3-CI algorithm for chl *a* (https://oceancolor.gsfc.nasa.gov/atbd/chlor_a/; accessed 15 December 2020). Coincident 4 km daily composites of SST (NASA OBPG, 2015) and photosynthetically active radiation (PAR) (NASA OBPG, 2017b) were also downloaded from the NASA ocean color website (<https://oceancolor.gsfc.nasa.gov/>; accessed 10 December 2020). SST, PAR and chl *a* were used to estimate net primary production (NPP) using the Wavelength Resolving Model (Morel, 1991) with the look up table described in Smyth et al. (2005). Daily net community production (NCP) composites were generated using NPP and SST data with the algorithm NCP-D described in Tilstone et al. (2015). The chl *a*, NPP, NCP and SST satellite algorithms were shown to perform best in the South Atlantic with respect to in situ data and accounting for uncertainties in both in situ and satellite data (Ford et al., 2021). Daily 8 km sea surface salinity (SSS) composites were retrieved from the Copernicus Marine Service (CMEMS) physics reanalysis product (GLORYS12) (CMEMS, 2021). Daily 0.25° resolution wind speed at 10 m were downloaded from Remote Sensing Systems Cross-Calibrated Multi-Platform (CCMP) product (Wentz et al., 2015). All data were retrieved for the period July 2002 to December 2018.

2.2. AVISO+ Mesoscale Eddy Tracking Product and Lagrangian Tracking

The satellite altimetry AVISO+ Mesoscale Eddy Product META3.1exp (Mason et al., 2014; Pegliasco et al., 2021, 2022) was used to identify the trajectories of mesoscale eddies within the South Atlantic Ocean, and provides daily estimates of the eddy location and radius. Anticyclonic (Agulhas) eddies were analyzed if: (a)

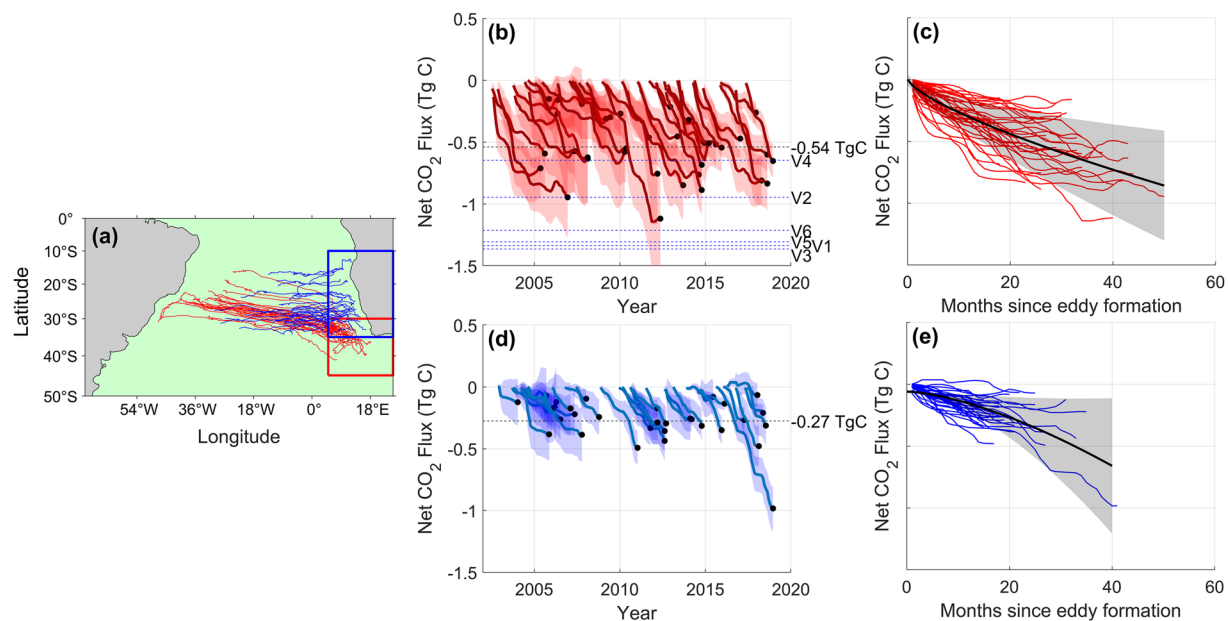


Figure 1. (a) Trajectories of the 36 anticyclonic (red lines) and 31 cyclonic (blue lines) eddies. Red and blue boxes indicate the formation region for the anticyclonic and cyclonic eddies respectively. (b) Red lines indicate the cumulative net CO₂ flux for the 36 anticyclonic eddies, where shading is the propagated uncertainty. Black dots indicate the cumulative net CO₂ flux at eddy dissipation. Black dashed line indicates the median cumulative net CO₂ flux at eddy dissipation (i.e., median of black dots). Blue dashed lines indicate the estimates for 6 anticyclonic eddies presented in Orselli, Kerr, et al. (2019). (c) Cumulative net CO₂ flux for the 36 anticyclonic eddies plotted since eddy formation. Black line indicates a power law fit ($y = a * x^b + c$) for the temporal evolution of the net CO₂ flux of the 36 anticyclonic eddies, where shading indicates the 95% confidence limits. (d) The same as (b), and (e) the same as (c) but for the 31 cyclonic eddies.

the eddy trajectory started in the Agulhas retroflection region (30°S–40°S; 5°E–25°E; Figure 1a); (b) the eddy trajectory was longer than 1 year; and (c) the trajectory crossed 0°E into the South Atlantic gyre region. These criteria identified 36 anticyclonic eddies for analysis between July 2002 and December 2018, that entered the South Atlantic as a single trajectory from formation to dissipation, with limited interactions with other eddies. The same selection procedure identified 31 cyclonic eddies originating from the Benguela upwelling system (15°S–40°S; 5°E–25°E; Figure 1a) for further analysis.

For each eddy, a timeseries of daily SST, SSS, NCP and wind speed was constructed using the eddy location and radius estimates from the AVISO+ product. For each parameter, the available data were extracted assuming a circular eddy and the median value taken when at least 30% of the data within the eddy were available. An example of the timeseries is given in Figure S1 in Supporting Information S1. To quantify the effect of eddies with respect to the surrounding conditions, daily timeseries of the environmental conditions were also extracted from a circular region three times the radius of the eddy (radii from two to five were tested; Figure S5 in Supporting Information S1), excluding data inside the eddy radius. Median monthly SST, SSS, NCP and wind speeds were calculated from daily timeseries, both for the eddy and the surrounding environment.

2.3. Sea Surface pCO₂ Estimates

The sea surface pCO₂ ($p\text{CO}_{2(\text{sw})}$) was determined for each month of the eddy trajectories using the South Atlantic Feed Forward Neural Network approach (SA-FNN_{NCP}; Ford et al. (2022)). The SA-FNN_{NCP} estimates $p\text{CO}_{2(\text{sw})}$ at the base of the mass boundary layer (sub skin $p\text{CO}_{2(\text{sw})}$) (Woelf et al., 2016) using non-linear relationships between $p\text{CO}_{2(\text{sw})}$ and three environmental drivers; $p\text{CO}_{2(\text{atm})}$, SST and NCP, which were constructed for eight static provinces in the South Atlantic Ocean. The SA-FNN_{NCP} was supplied with the monthly median SST and NCP, and the $p\text{CO}_{2(\text{atm})}$ for the mean location of the eddy within the month. $p\text{CO}_{2(\text{atm})}$ was estimated using the dry mixing ratio of CO₂ from the National Oceanic and Atmospheric Administration Earth System Research Laboratories (NOAA-ESRL) marine boundary layer reference, skin SST and sea level pressure following Dickson et al. (2007). The $p\text{CO}_{2(\text{sw})}$ uncertainty was estimated by propagating the $p\text{CO}_{2(\text{atm})}$ (1 μatm), satellite SST (0.441°C) and NCP (45 mmol O₂ m⁻² d⁻¹) (Ford et al., 2021) uncertainties through the SA-FNN_{NCP}, and

combined in quadrature with the SA-FNN_{NCP} uncertainty (21.48 μatm) (Ford et al., 2022) using standard uncertainty propagation methods (Taylor, 1997).

2.4. Estimation of the Cumulative Bulk Air-Sea CO₂ flux

The air-sea CO₂ flux (F) was calculated for each month of the eddy trajectory using a bulk parameterization as:

$$F = k (\alpha_w p\text{CO}_{2(\text{sw})} - \alpha_s p\text{CO}_{2(\text{atm})}) \quad (1)$$

where k is the gas transfer velocity estimated from median wind speeds following the parameterization of Nightingale et al. (2000). α_w and α_s are the solubility of CO₂ at the base and top of the mass boundary layer at the sea surface (Woolf et al., 2016). α_w was calculated as a function of the skin SST and SSS (Weiss, 1974), applying a cool skin bias of +0.17 K to convert the skin SST to sub skin SST (Donlon et al., 1999; Woolf et al., 2016). α_s was calculated as a function of the eddy skin SST and the SSS with a salinity gradient of +0.1 salinity units between the base and top of the mass boundary layer (Woolf et al., 2016). The CO₂ flux calculations were carried out with the open source FluxEngine toolbox (Holding et al., 2019; Shutler et al., 2016) using the “rapid transport” approximation (described in Woolf et al., 2016) at monthly time steps.

The monthly average daily flux of CO₂ ($\text{mmol C m}^{-2} \text{d}^{-1}$) was multiplied by the number of days and the area of the eddy, assuming a circular eddy with the mean eddy radius, in the respective month. The fluxes (Tg C mon^{-1}) were then added cumulatively to retrieve the net cumulative CO₂ flux for each eddy. The uncertainties in $p\text{CO}_{2(\text{sw})}$ (temporally varying), $p\text{CO}_{2(\text{atm})}$ (1 μatm), SST (0.441°C) (Ford et al., 2021) and the gas transfer velocity (assumed to be $\pm 10\%$; Woolf et al., 2019) were propagated through the cumulative flux calculations using a Monte Carlo uncertainty propagation ($N = 1000$), and the 95% confidence interval (2 standard deviations) extracted as the uncertainty on the cumulative net CO₂ flux. These cumulative CO₂ flux calculations were repeated for the surrounding environment conditions, assuming the same area as the eddy to estimate the cumulative CO₂ flux as if the eddy were not present. A Mann Whitney U-Test was used to assess whether the median percentage differences between the eddy and surrounding environment CO₂ flux were significantly different from 0. The Mann Whitney U-Test was selected due to the sample numbers within our study and the lower sensitivity of the test to outliers in the data.

2.5. Thermal and Non-thermal Decomposition of $p\text{CO}_{2(\text{sw})}$ Timeseries

The eddy $p\text{CO}_{2(\text{sw})}$ timeseries was separated into its thermal and non-thermal components as described in Takahashi et al. (2002), where we refer the reader for further details. In brief, the thermal component ($p\text{CO}_{2(\text{therm})}$) was calculated as:

$$p\text{CO}_{2(\text{therm})} = p\text{CO}_{2(\text{sw})} \times e^{(0.0423 \times (\overline{\text{SST}} - \text{SST}))} \quad (2)$$

$\overline{\text{SST}}$ and SST are the mean subskin SST across the eddy timeseries and the monthly subskin SST respectively. The non-thermal component ($p\text{CO}_{2(\text{bio})}$) was calculated as:

$$p\text{CO}_{2(\text{bio})} = \overline{p\text{CO}_{2(\text{sw})}} \times e^{(0.0423 \times (\text{SST} - \overline{\text{SST}}))} \quad (3)$$

$\overline{p\text{CO}_{2(\text{sw})}}$ was the mean $p\text{CO}_{2(\text{sw})}$ for the eddy timeseries. The contributions of the two competing components to the $p\text{CO}_{2(\text{sw})}$ timeseries can be determined from the seasonal amplitude of the $p\text{CO}_{2(\text{therm})}$ and $p\text{CO}_{2(\text{bio})}$:

$$\Delta p\text{CO}_{2(\text{therm})} = [p\text{CO}_{2(\text{therm})}]_{\text{max}} - [p\text{CO}_{2(\text{therm})}]_{\text{min}} \quad (4)$$

$$\Delta p\text{CO}_{2(\text{bio})} = [p\text{CO}_{2(\text{bio})}]_{\text{max}} - [p\text{CO}_{2(\text{bio})}]_{\text{min}} \quad (5)$$

The seasonal amplitudes were calculated using a 12-month moving window for the eddy lifetime, and the ratio between the thermal and non-thermal component (R) was determined as:

$$R = \frac{\Delta p\text{CO}_{2(\text{therm})}}{\Delta p\text{CO}_{2(\text{bio})}} \quad (6)$$

Table 1

The Calculation of the Modification to the South Atlantic Ocean CO₂ Sink That Mesoscale Eddies May Contribute

	Anticyclonic	Cyclonic	
Median cumulative CO ₂ flux (Figures 1b and 1d; Tg C per eddy)	−0.54 (IQR = 0.38)	−0.27 (IQR = 0.17)	
Median percentage change in CO ₂ flux compared to surrounding environment (Figure 2c; %)	−3.7 (IQR = 5.2)	−1.4 (IQR = 4.8)	
Additional flux into eddy (Tg C per eddy)	−0.020 ± 0.015	−0.004 ± 0.006	
Mean eddy lifetime (yr)	2.5	1.7	
Additional flux into eddy per year (Tg C yr ^{−1})	−0.008 ± 0.006	−0.002 ± 0.003	
Spawn Rate (yr)	6 (Lutjeharms, 2006)	4 (Chaigneau et al., 2009)	
Additional flux into eddies (Tg C yr ^{−1})	−0.05 ± 0.03	−0.01 ± 0.02	
			Total
South Atlantic Ocean CO ₂ sink estimate of Ford et al. (2022) (−76 ± 8 Tg C yr ^{−1}) 20°S–44°S	−0.06 ± 0.04%	−0.02 ± 0.02%	−0.08 ± 0.04%
South Atlantic Ocean CO ₂ sink estimate of Woolf et al. (2019) and Holding et al. (2019) (−261 ± 28 Tg C yr ^{−1}) 20°S–44°S	−0.020 ± 0.015%	−0.005 ± 0.008%	−0.030 ± 0.013%

Note. The median percentage change in the eddy flux compared to the surrounding environment is converted to a median Tg C yr^{−1} equivalent and compared to two estimates of the South Atlantic CO₂ sink in the region the eddies propagate. IQR is the interquartile range.

In cases where R is greater (less) than 1, the thermal (non-thermal) contribution was the dominant driver. The anomaly in R was determined by subtracting the mean R across the eddy's lifetime.

3. Results

The accurate estimation of $p\text{CO}_{2(\text{sw})}$ by the SA-FNN_{NCP} is an important component of the air-sea CO₂ flux calculation. A comparison between the SA-FNN_{NCP} estimated and in situ $p\text{CO}_{2(\text{sw})}$ within both anticyclonic ($n = 6$) and cyclonic eddies ($n = 2$; Figure S2 in Supporting Information S1) was therefore performed. The showed that the SA-FNN_{NCP} was accurate and precise within anticyclonic eddies (root mean square deviation = 10 μatm ; bias = 0 μatm) but larger differences were apparent in $p\text{CO}_{2(\text{sw})}$ for the cyclonic eddies, albeit from just two cross-overs (root mean square deviation = 21 μatm ; bias 11 μatm). The comparison provides confidence in the resulting air-sea CO₂ flux estimates.

The analysis of the eddy cumulative CO₂ flux showed both anticyclonic (Agulhas; Table 1) and cyclonic (Benguela; Table 1) eddies acted as net CO₂ sinks over their lifetime (Figures 1b and 1d). Anticyclonic eddies displayed an exponential decay in the increase of the net cumulative CO₂ sink, compared to a more linear increase in cyclonic eddies using the same functional equation (Figures 1c and 1e).

The anomaly in the thermal to non-thermal contribution to $p\text{CO}_{2(\text{sw})}$ variability in anticyclonic eddies changed over their lifetimes (Figure 2a), where a positive anomaly indicates an increasing dominance of temperature on controlling $p\text{CO}_{2(\text{sw})}$. For cyclonic eddies the anomaly in the thermal to non-thermal component ratio, did not change significantly over time (Figure 2b).

The anticyclonic (−3.7%, Mann-Whitney U-Test, $p < 0.001$, $n = 36$) and cyclonic (−1.4%, Mann-Whitney U-Test, $p = 0.007$, $n = 31$) eddies significantly enhanced the cumulative CO₂ sink compared to the water surrounding each eddy (Figure 2c). No significant differences in this enhancement were observed between anticyclonic and cyclonic eddies (Figure 2c; Mann-Whitney U-Test, $p = 0.16$), although the median anticyclonic modification was double that of the cyclonic eddies (Figure 2c).

4. Discussion

To our knowledge, this is the first study to provide an observation-based assessment of the cumulative net CO₂ flux of a large number (67) of long-lived mesoscale eddies. The trajectory of 36 anticyclonic eddies was followed over their lifetime and showed that they were a net cumulative CO₂ sink (median = 0.54 Tg C per eddy; Table 1). This result is consistent with the lower end of values from the literature for anticyclonic (Agulhas) eddies which

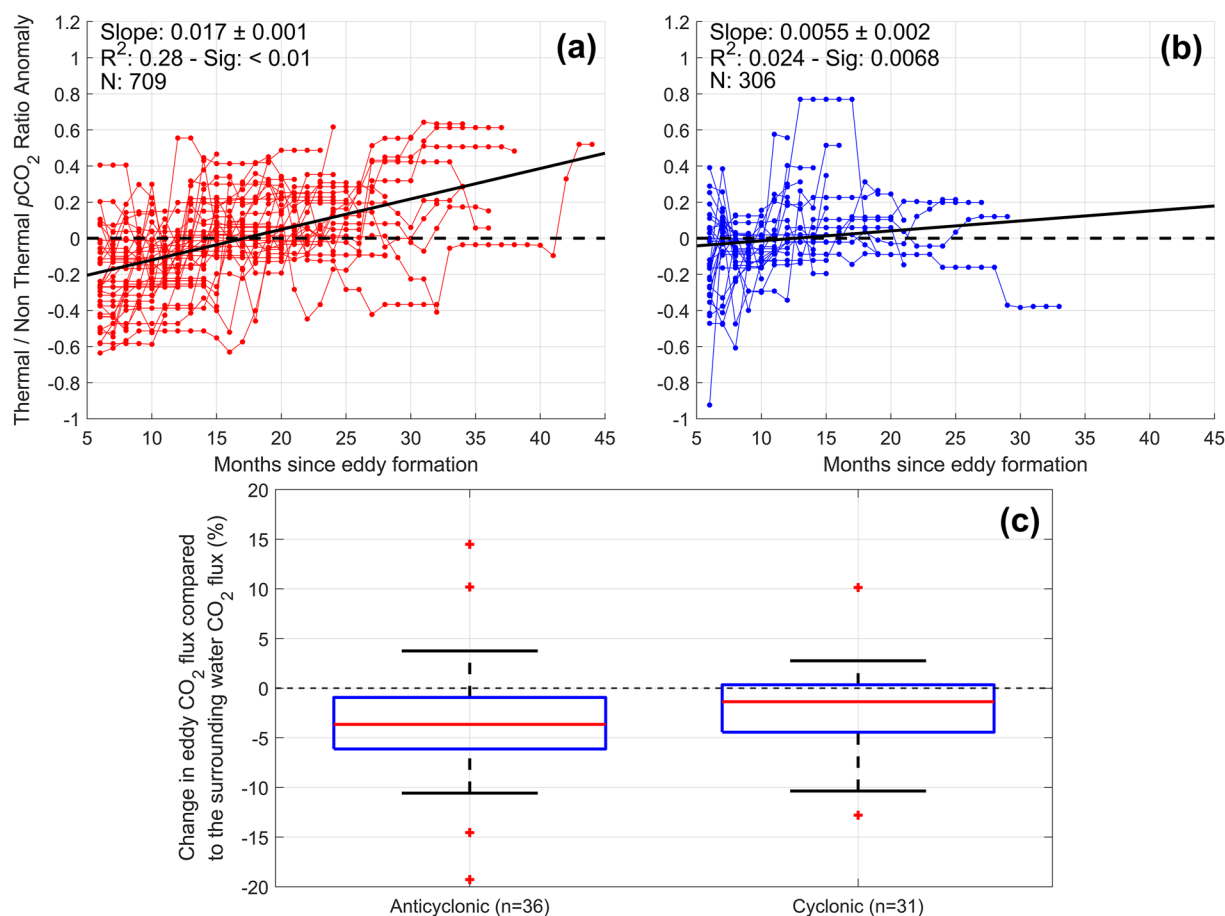


Figure 2. (a) Anomaly in the 12-month running thermal to the non-thermal ratio of $p\text{CO}_{2(\text{sw})}$ for the 36 anticyclonic eddies. Black solid line indicates the linear fit since the formation of the eddy. Black dashed line indicates an anomaly of 0. Statistics within the plot are: Slope is the slope of the linear fit, R^2 is the coefficient of determination, Sig is the significance of the linear fit and N is number of samples. (b) Same as (a) but for the 31 cyclonic eddies. (c) Box plots indicating the percent change in the cumulative net CO_2 flux at eddy dissipation with respect to the waters surrounding the eddy. Red line indicates the median, blue box indicates the 25th and 75th percentile and whiskers show the minimum and maximum non-outlier values. Red crosses indicate outliers that are more than 1.5 times the interquartile range from the 25th and 75th percentiles. Negative percentages indicate a stronger flux, where positive percentages indicate a weaker flux.

have identified them as a net sink for CO_2 varying from ~ 0.6 to ~ 1.4 Tg C (median = 1.26 Tg C per eddy; Figure 1b; Orselli, Kerr, et al., 2019). The results of the Orselli, Kerr, et al. (2019) study were based on extrapolation of a snapshot of the eddies CO_2 uptake potential from ship observations that crossed the paths of six eddies. Two of these eddies were tracked in our study (V1, V3; Figure 1b) and identified as CO_2 sinks of 0.64 and 0.40 Tg C compared to 1.34 and 1.36 Tg C by Orselli, Kerr, et al. (2019). The eddies sampled by Orselli, Kerr, et al. (2019) were in austral winter when they were a strong CO_2 sink. In our study, the seasonal variability in the CO_2 flux is captured, where eddies were stronger sinks in winter and weaker sinks for CO_2 in summer, which likely explains the lower cumulative CO_2 flux. This seasonality in the CO_2 flux would partially be due to seasonal SST and wind speed cycles, where cooler SST increases CO_2 solubility (e.g., Figures S1a and S1b in Supporting Information S1) and elevated wind speeds increase gas transfer during winter (Kitidis et al., 2019).

Both the anticyclonic and cyclonic eddies showed an increasing cumulative CO_2 sink over their lifetime (Figures 1c and 1e). For the former, the rate of CO_2 uptake decreased exponentially over this period (Figure 1c). This result is consistent with the geographical propagation of the eddies in the oligotrophic South Atlantic gyre (Figure 1a), and eddy stirring of the environment (McGillicuddy, 2016). The significant increase in the anomalies of the seasonal thermal to non-thermal $p\text{CO}_{2(\text{sw})}$ ratio (becoming more influenced by temperature; Figure 2a) was found to be mainly driven by a relative reduction in the non-thermal contribution (not shown) and highlights the changing role of biological activity and/or circulation over time as the eddies propagated into the gyre. Carvalho et al. (2019) showed that the phytoplankton community structure changed as the eddies aged, where younger

anticyclonic (Agulhas) eddies were dominated by haptophytes (small flagellates) followed by prokaryotes. The shift in community structure was attributed to a decrease in nutrient availability at the surface, whereby prokaryotes are better adapted to lower nutrient concentrations. Sarkar et al. (2021) highlighted that haptophytes are crucial for the biological CO₂ drawdown in the Agulhas retroflection, reinforcing a weaker biological pump as the eddies evolve. In contrast, the cyclonic eddies displayed a linear increase in the cumulative CO₂ sink (Figure 1e). This signature may be because the cyclonic eddies do not propagate as far as anticyclonic eddies into the South Atlantic gyre (Figure 1a), which is also illustrated by no significant change in the thermal to non-thermal $p\text{CO}_{2(\text{sw})}$ ratio anomaly (Figure 2b).

Both the anticyclonic and cyclonic eddies were shown to significantly increase the CO₂ drawdown in the South Atlantic Ocean (Figure 2c), compared to the surrounding environment. Jones et al. (2017) showed that both anticyclonic and cyclonic eddies were hotspots for CO₂ drawdown in the Antarctic Circumpolar Current. Dufois et al. (2016) examined chl *a* variability in anticyclonic eddies and showed that the first two modes of spatial variability were consistent with eddy stirring, and the third mode highlighted the mesoscale modification. In our study, by comparing the cumulative CO₂ fluxes of the eddies to a theoretical eddy consisting of surface waters surrounding the eddy, the mesoscale modulation of the air-sea CO₂ flux was quantified. We showed this modulation to increase the CO₂ sink into anticyclonic and cyclonic eddies by 3.7% and 1.4% respectively (Figure 2c).

The cyclonic eddies generally showed lower SST, and higher NCP (Figures S3 and S4 in Supporting Information S1) compared to the surrounding waters, suggesting that both biological and physical processes amplify the CO₂ sink. Chen et al. (2007) showed in the North Pacific that $p\text{CO}_{2(\text{sw})}$ was elevated at the core of a cyclonic eddy, due to the upwelling of CO₂ rich waters into the surface layer and the eddy acting as a weaker CO₂ sink compared to the surrounding waters. By comparison, Lovecchio et al. (2022) identified that cyclonic eddies around the Canary upwelling system entrain nearshore nutrient rich waters into the eddy core at formation. Mesoscale upwelling of nutrients was a small component of the total nutrients sustaining the biological production. This suggests that the biological CO₂ drawdown throughout the eddy lifetime is largely supported by the initial nutrient input which ultimately enhances the CO₂ sink through both the physical and biological signatures.

The anticyclonic eddies were associated with elevated SST at formation, which rapidly changed to depressed SST for the remainder of their lifetimes (Figure S3 in Supporting Information S1) compared to the surrounding waters. NCP remained lower than the surroundings (Figure S4 in Supporting Information S1). These characteristics suggest opposing physical and biological forces that modify the air-sea CO₂ flux. Similarly, Laxenaire et al. (2019) showed that the SST anomaly associated with surface water of an anticyclonic (Agulhas) eddy switched from positive to negative over its lifetime, also implying a change from a CO₂ source to sink as it propagated over the South Atlantic basin. This indicates that the physical component exerts the greatest control on amplifying the air-sea CO₂ sink into these anticyclonic eddies. The magnitude of SST differences in anticyclonic eddies compared to the surrounding environment were generally double that of cyclonic eddies which is indicative of the mechanism by which anticyclonic eddies enhance the CO₂ sink more than cyclonic eddies.

This study has identified the capacity for eddies to modify the air-sea CO₂ flux (Figure 2c) which is driven by intrinsic differences between individual eddies (Figures S3 and S4 in Supporting Information S1). Lehahn et al. (2011) observed an isolated patch of elevated chl *a* associated with an anticyclonic eddy that was transported into the South Atlantic gyre, suggesting enhanced biological drawdown of CO₂, though it was not possible to identify if this is a common feature of all anticyclonic eddies. Entrainment of nutrient rich nearshore waters into the cyclonic eddies (Lovecchio et al., 2022) is likely to be highly variable depending on the location and interaction with other water bodies and the time of year, which will in turn lead to a different biological response and therefore air-sea CO₂ flux. Many mesoscale eddy studies are limited by the availability of in situ data (Jones et al., 2017; Laxenaire et al., 2019; Orselli, Kerr, et al., 2019). The expanding use of Biogeochemical-Argo profilers, especially those with pH sensors (Roemmich et al., 2019), is improving the potential to assess the air-sea CO₂ flux both globally and regionally (Gray et al., 2018). A synergy of in situ and satellite observations is the most comprehensive means of studying the processes by which mesoscale eddies modify the air-sea CO₂ flux.

Based on a recent assessment of the South Atlantic Ocean (20°S–44°S) by Ford et al. (2022), which estimated the region to be a CO₂ sink of 76 Tg C yr^{−1}, the long-lived anticyclonic (Agulhas) eddies assuming six eddies are released per year (Lutjeharms, 2006) would contribute 1.3 Tg C yr^{−1} (1.7%; Table 1). Orselli, Kerr, et al. (2019) identified that six anticyclonic (Agulhas) eddies contributed 2.5 Tg C yr^{−1} (3.3%) to the CO₂ sink. Our contribution is lower because seasonal variability in the CO₂ flux was captured.

We found that anticyclonic and cyclonic eddies enhance the oceanic CO₂ sink into the South Atlantic Ocean (20°S–44°S) by $0.08 \pm 0.04\%$ (Table 1). Globally, long lived mesoscale eddies, make up 0.4% of the eddy trajectories in the AVISO+ data set (Pegliasco et al., 2022). This suggests that the effect of all eddies on the CO₂ flux and net oceanic sink is likely to be globally significant. Eddy kinetic energy, as a proxy for mesoscale eddy occurrence, has been increasing at a rate between 2% and 5% per decade (Martínez-Moreno et al., 2021), indicating that the role of mesoscale eddies on the oceanic CO₂ sink may be becoming more significant. In the context of climate change and increasing global temperatures, further work is required to quantify the influence of these changes on the ocean CO₂ sink.

5. Conclusions

Our analysis presents a novel approach to assess the impact of long-lived mesoscale eddies on the air-sea CO₂ flux in the South Atlantic Ocean. Using satellite observations, in situ data and Lagrangian tracking we show that anticyclonic and cyclonic eddies are cumulative net CO₂ sinks of 0.54 and 0.27 Tg C per eddy (median values), respectively. Anticyclonic eddies exhibited an exponential decay in the rate of CO₂ uptake, and significant changes in the thermal to non-thermal drivers of the $p\text{CO}_{2(\text{sw})}$ ratio anomaly. This shows that the thermal and biological contributions to the CO₂ sink variability change as the eddies age and propagate over different geographic trajectories in the South Atlantic gyre. The cyclonic eddies showed a more linear rate of CO₂ uptake, and there was no significant change in the drivers of the seasonal $p\text{CO}_{2(\text{sw})}$ ratio anomaly.

Both anticyclonic and cyclonic eddies amplified the CO₂ sink compared to the surrounding environment by 3.7% and 1.4%, respectively. For the anticyclonic eddies, physical drivers increased the CO₂ sink, whereas the biological component reduced the uptake. In cyclonic eddies both physical and biological components worked synergistically to increase the CO₂ sink. Accounting for their typical frequencies, long-lived mesoscale eddies significantly amplify the CO₂ sink into the South Atlantic Ocean (20°S–44°S) by $0.08 \pm 0.04\%$. Although this modification appears small, long-lived eddies make up 0.4% of eddies in the global oceans, and therefore the amplification of the global CO₂ sink from all eddies is likely to be much larger than previously estimated. The inclusion of these mesoscale features would improve estimates of the role of the global ocean in the uptake of anthropogenic CO₂.

Data Availability Statement

Daily Moderate Resolution Imaging Spectroradiometer on Aqua (MODIS-A) estimates of chlorophyll-a (NASA OBPG, 2017a), photosynthetically active radiation (NASA OBPG, 2017b) and sea surface temperature (NASA OBPG, 2015) are available from the National Aeronautics Space Administration (NASA) ocean color website (<https://oceancolor.gsfc.nasa.gov/>). Modeled sea surface salinity from the Copernicus Marine Environment Modelling Service (CMEMS) global ocean physics reanalysis product (GLORYS12V1) are available from CMEMS (CMEMS, 2021). The CCMP daily wind speed products are available from Remote Sensing Systems (www.remss.com/measurements/ccmp; Wentz et al., 2015). In situ GO-SHIP DIC and TA samples can be downloaded from the NOAA/NODC data centre (<https://www.ncei.noaa.gov/access/ocean-carbon-data-system/oceans/RepeatSections/>). FORSA in situ $p\text{CO}_{2(\text{sw})}$ data can be retrieved from PANGAEA (Orselli et al., 2023). Optimum Interpolated SST (OISST) v2 (Reynolds et al., 2002) data used in the reanalysis of $p\text{CO}_{2(\text{sw})}$ can be downloaded from <https://psl.noaa.gov/data/gridded/data.noaa.oisst.v2.html>. The AVISO+ Mesoscale Eddy Product META3.1exp can be downloaded from <https://doi.org/10.24400/527896/a01-2021.001> (Pegliasco et al., 2021).

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