

# Contrasting trends of the ocean CO<sub>2</sub> sink and pH in the agulhas current system and the Mozambique basin, south-western Indian ocean (1963–2023)<sup>☆</sup>

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## ABSTRACT

We describe new observations of the oceanic carbonate system in the South-Western Indian Ocean obtained in January 2021 (OISO-31 cruise) and May 2022 (RESILIENCE cruise). To evaluate the decadal trends and drivers of fugacity of CO<sub>2</sub> (fCO<sub>2</sub>), air-sea CO<sub>2</sub> fluxes, dissolved inorganic carbon (C<sub>T</sub>) and pH, we used available data in this region over 1963–2023 and compared the results in the Mozambique Basin and in the Agulhas region near the African coast. Over 1995–2023, we found a faster fCO<sub>2</sub> increase in the Mozambique basin ( $2.03 \pm 0.07 \text{ \mu atm.yr}^{-1}$ ) compared to the coastal zone ( $1.37 \pm 0.07 \text{ \mu atm.yr}^{-1}$ ). The temporal change of anthropogenic CO<sub>2</sub> concentrations estimated in subsurface enables to reconstruct the carbonate system properties since the 1960s. In the Mozambique Basin the CO<sub>2</sub> sink increased slightly over 1960–2022 with a maximum observed in May 2022 ( $-2.4 \text{ mmolC.m}^{-2}.\text{d}^{-1}$ ). In the coastal zone, the ocean CO<sub>2</sub> sink increased from near equilibrium in the 1960s to a maximum observed in May 2022 ( $-4.2 \text{ mmolC.m}^{-2}.\text{d}^{-1}$ ). In both regions, we found a decrease of pH, most pronounced in the open ocean zone ( $-0.020 \pm 0.001.\text{decade}^{-1}$  over 1995–2023). The lowest pH of 8.04 was observed in January 2021, 0.11 lower than in the 1960s. The increase of the CO<sub>2</sub> sink and the decrease of pH were mainly driven by anthropogenic CO<sub>2</sub> uptake, with about 10% due to the ocean warming.

## 1. Introduction

The ocean plays a major role in reducing the impact of climate change by absorbing more than 90% of the excess heat in the climate system (Cheng et al., 2020) and about 25% of human released CO<sub>2</sub> (Friedlingstein et al., 2022, 2023). The oceanic CO<sub>2</sub> uptake also changes the chemistry of seawater reducing its buffering capacity (Revelle and Suess, 1957) and leading to a process known as ocean acidification (OA) with potential impacts on marine organisms (Fabry et al., 2008; Doney et al., 2009, 2020). Although important international effort have been conducted to synthesize fCO<sub>2</sub> observations (Bakker et al., 2016) and their use to reconstruct fCO<sub>2</sub> fields and air-sea CO<sub>2</sub> fluxes (Rödenbeck et al., 2015), there are still large uncertainties on the ocean CO<sub>2</sub> sink estimate at global scale (De Vries et al., 2023; Friedlingstein et al., 2022,

2023) and in coastal zones (Resplandy et al., 2024). At seasonal scale large differences between observations and models were identified leading to difference of the global coastal ocean CO<sub>2</sub> sink up to 60% (Resplandy et al., 2024). To reduce these uncertainties, it is thus important to better document the carbonate system (CS) for comparing and correct Ocean Biogeochemical Global Models (OBGM) and climate/carbon models (CMIP) used to predict future changes of the ocean states including ocean acidification (e.g. Jiang et al., 2023) and their impact on the marine ecosystems (Doney et al., 2020; Kwiatkowski et al., 2020). Coastal waters experienced enhanced ocean acidification due to increase CO<sub>2</sub> uptake (Bourgeois et al., 2016; Laruelle et al., 2018; Roobaert et al., 2024). As opposed to open ocean zones, it is difficult to detect acidification trends in the heterogeneous coastal waters due to combined effects of anthropogenic CO<sub>2</sub>, ocean warming, local

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anthropogenic inputs through rivers and from air pollution (e.g. [Sarma et al., 2015](#); [Sridevi and Sarma, 2021](#)), as well as due to variability of upwelling and biological processes like observed in the Western Boundary Current (WBC) such as the Agulhas Current system investigated here. In this region, identified as MARCATS #25 (Margins and CATchment Segmentation, [Laruelle et al., 2013](#)), only one study evaluated the CO<sub>2</sub> fluxes based on fCO<sub>2</sub> data ([Arnone et al., 2017](#)). As noted by these authors, 8 years of data (in 2005–2012) enabled to estimate the mean flux in this region, a CO<sub>2</sub> sink of  $-1.65 \pm 0.04 \text{ molC.m}^{-2}.\text{yr}^{-1}$ , but cannot separate the natural decadal and anthropogenic signal. Concerning the ocean acidification in African coastal zones observations were only available in the western side ([González-Dávila et al., 2017](#)): these authors estimated a pH decrease of  $-0.03 \pm 0.01.\text{decade}^{-1}$  but this was evaluated for a short period (2005–2012). Based on fCO<sub>2</sub> data and using Alkalinity/Salinity (A<sub>T</sub>/S) relationship, [Lauvset et al. \(2015\)](#) estimated a pH trend in the Indian Ocean Subtropical biome (STPS) of  $-0.024 \pm 0.004.\text{decade}^{-1}$  for the period 1981–2011. Recently, reconstructed sea surface carbonate system properties for the period 1985–2021 suggested pH trend of between  $-0.020$  and  $-0.017.\text{decade}^{-1}$  in the South-Western Indian Ocean ([Chau et al., 2024](#)). This analysis, along with other data-products and ocean models was then used to quantify the trends and drivers of the ocean acidification in the Indian Ocean ([Chakraborty et al., 2024](#)). These authors highlighted an acceleration of the pH trend from  $-0.011 (\pm 0.00).\text{decade}^{-1}$  in 1980–1989 to  $-0.019 (\pm 0.004).\text{decade}^{-1}$  in 2010–2019. To investigate the long-term changes of pH in the Indian Ocean [Madkaiker et al. \(2023\)](#) also used outputs from ocean models and found that over 1961–2010 the decrease of pH was mainly driven by the increase of dissolved inorganic carbon (69%) with a significant contribution of the warming (14%). In the Mozambique Channel, [Lo Monaco et al. \(2021\)](#) estimated a pH decrease of  $-0.023 \pm 0.005.\text{decade}^{-1}$  over 1995–2019. These results highlight the contrasting pH trend between open ocean and coastal zones and depending the period. In a recent review of the biogeochemistry in the Indian Ocean, [Ghosh et al. \(2024\)](#) concluded that “research on ocean acidification in the Indian Ocean is in its early

stages” and that “coordinated multidisciplinary chemical and biological observational approaches are needed to document the status of ocean acidification in the high seas and coastal environments”. In this context we present new observations obtained in 2021 and 2022 as part of the OISO-31 and RESILIENCE cruises conducted in the South Western Indian Ocean, the Mozambique Basin, the Agulhas current system and African coastal zone. To investigate the long-term change of the air-sea CO<sub>2</sub> fluxes and of the carbonate system we also used historical data in this region obtained over 1963–2023.

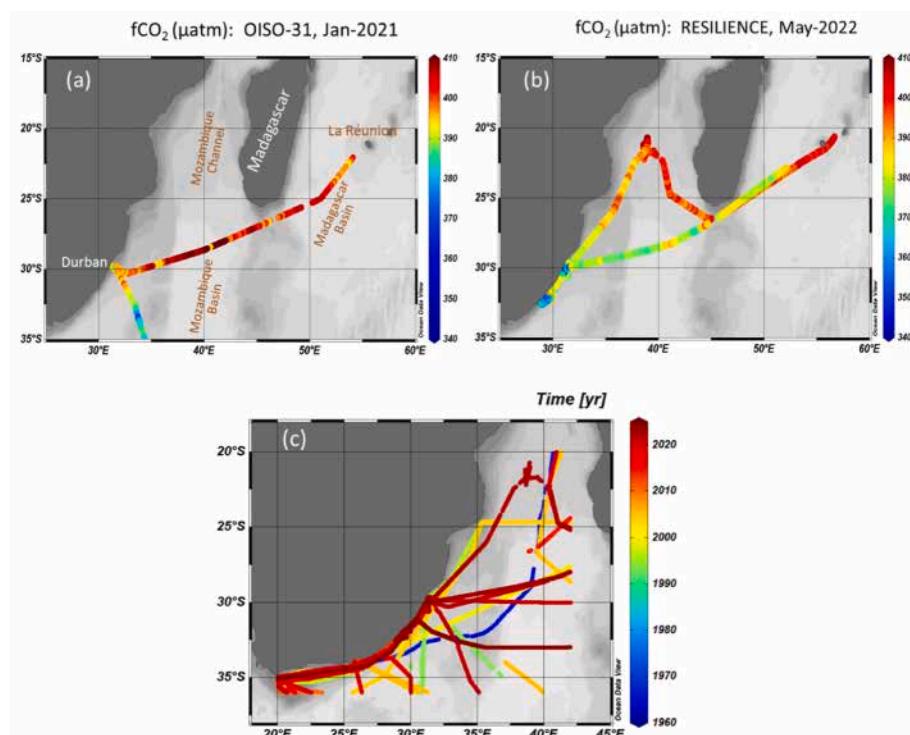
## 2. Data selection and methods

### 2.1. Data selection

The OISO-31 (January–March 2021) and RESILIENCE (May 2022) cruises were conducted in the South-Western Indian Ocean. Part of these cruises followed the same track in the Madagascar and the Mozambique Basins (Fig. 1). To explore the long-term change of the carbonate system in this region, we used the SOCAT data, version v2024 ([Bakker et al., 2016, 2024](#), Fig. 1c, Supp. Mat. Table S1a) and the GLODAP data, version v2023 ([Lauvset et al., 2024](#), Supp. Mat. Table S1b). As we used salinity in the carbonate system calculations and salinity was not quality controlled in SOCAT, we have checked the salinity and found bias for few cruises (in 2011 and 2012); for these cruises we thus used the World Ocean Atlas (WOA) salinity as listed in the SOCAT data files ([Antonov et al., 2006](#); [Pfeil et al., 2013](#)).

### 2.2. Methods

The methods for surface underway fCO<sub>2</sub> and biogeochemical properties (Oxygen, A<sub>T</sub>, C<sub>T</sub>, nutrients) were described in previous studies (e.g. [Lo Monaco et al., 2021](#); [Metzl et al., 2022](#)). Here we briefly recall the methods for underway fCO<sub>2</sub> and water-column observations.



**Fig. 1.** Maps of OISO-31 cruise (a) and RESILIENCE cruise (b) conducted in January 2021 and April–May 2022; color code is for sea surface fCO<sub>2</sub> (μatm). (c) Tracks of cruises in the SOCAT data-base, v2024 ([Bakker et al., 2016, 2024](#)); color code is for Year (1963–2023). Figures produced with ODV ([Schlitzer, 2018](#)).

### 2.2.1. Surface $f\text{CO}_2$ and $A_T$ $C_T$ data

For  $f\text{CO}_2$  measurements during OISO-31 (2021) and RESILIENCE (2022) cruises, sea-surface water was continuously equilibrated with a "thin film" type equilibrator thermostated with surface seawater (Poisson et al., 1993) and  $x\text{CO}_2$  in the dried gas was measured with a non-dispersive infrared analyzer (NDIR, Siemens Ultramat 6F). Standard gases for calibration (279.31 ppm or 267.39 ppm, 350.75 ppm or 374.92 ppm and 489.09 ppm) were measured every 6 h. The  $f\text{CO}_2$  in situ data were corrected for warming using corrections proposed by Copin-Montégut (1988, 1989). Note that when incorporated in the SOCAT data-base, the original  $f\text{CO}_2$  data are recomputed (Pfeil et al., 2013) using temperature correction from Takahashi et al. (1993). Given the very small difference between equilibrium temperature and sea surface temperature ( $+0.088 \pm 0.066$ ,  $n = 6416$ , on average for the RESILIENCE cruise in 2022), the  $f\text{CO}_2$  data from SOCAT used in this analysis (Bakker et al., 2024) are almost identical (within 1  $\mu\text{atm}$ ) to the original  $f\text{CO}_2$  values (Lo Monaco and Metzl, 2024).

Total alkalinity ( $A_T$ ) and dissolved inorganic carbon ( $C_T$ ) were measured continuously in surface water (3–4 sample/hour) using a potentiometric titration method (Edmond, 1970) in a closed cell. For calibration, we used the Certified Referenced Materials (CRMs, Batch #182 and 191 for OISO-31) provided by Pr. A. Dickson (SIO, University of California). The repeatability of  $A_T$  and  $C_T$  measurements was evaluated from duplicate analyses of continuous sea surface sampling at the same location (when the ship was stopped). The mean differences of 343 duplicates were better than 3  $\mu\text{mol kg}^{-1}$  for  $A_T$  and  $C_T$  (Supp. Mat. Table S2). We estimated the accuracy for both  $A_T$  and  $C_T$  better than 3  $\mu\text{mol kg}^{-1}$ . The  $A_T$  and  $C_T$  data from OISO-31 cruise are available at NCEI/OCADS (Lo Monaco et al., 2023). These data offered comparisons and validation for the calculations of the carbonates properties using  $f\text{CO}_2$  data and  $A_T$ /Salinity relationship.

### 2.2.2. Carbonate system calculation and $A_T$ /salinity relationship

When two of the carbonate system properties are measured ( $f\text{CO}_2$ ,  $A_T$ ,  $C_T$  and pH) they can be used to calculate other species including the saturation state of aragonite and calcite ( $\Omega_{\text{Ar}}$ ,  $\Omega_{\text{Ca}}$ ). Here we have measurements of either  $f\text{CO}_2$  underway data or  $A_T$  and  $C_T$  from both underway sampling and in the water column at stations. To calculate pH and  $C_T$  from the  $f\text{CO}_2$  data we used the CO2sys program (version CO2sys\_v2.5, Orr et al., 2018) developed by Lewis and Wallace (1998) and adapted by Pierrot et al. (2006) with K1 and K2 dissociation constants from Lueker et al. (2000) and KSO4 constant from Dickson (1990). The total boron concentration is calculated according to Uppström (1974). When using  $f\text{CO}_2$  data to derive pH or  $C_T$ , one needs  $A_T$  concentrations that can be derived from salinity (e.g. Millero et al., 1998; Lauvset et al., 2015; Lo Monaco et al., 2021). Here we used the underway surface  $A_T$  observations obtained in 2004 (OISO-11) and 2021 (OISO-31) and compared the  $A_T$ /Salinity relationship with that derived from stations data in the surface layer (0–20m) in 1987–2021 in this region (Fig. 2). We thus obtained two relationships:

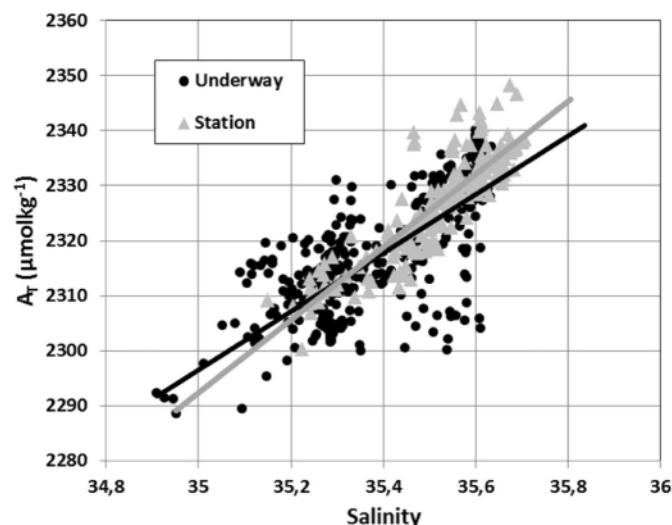
From underway data:

$$A_T (\mu\text{mol.kg}^{-1}) = 53.2626 (\pm 2.23) * \text{Salinity} + 432.359 (\pm 78.79) \quad (n = 408, r^2 = 0.58) \quad (\text{Eq. (1)})$$

From Stations data:

$$A_T (\mu\text{mol.kg}^{-1}) = 66.1773 (\pm 2.55) * \text{Salinity} - 23.8222 (\pm 90.55) \quad (n = 205, r^2 = 0.77) \quad (\text{Eq. (2)})$$

Both relationships lead to the same results for the  $C_T$  or pH calculations and we used the relationship from the stations (Eq. (2)) assuming there is no long-term change in salinity and alkalinity in this region. The validation of the calculation with underway surface measurements in 2021 is shown in Fig. S1 (Table S3). Given the errors in the CO2sys calculations (errors on constants K1, K2 and measurements, Orr et al., 2018) we consider the  $A_T$ /Salinity relationship valid to calculate accurately the  $C_T$  concentrations and pH in this region. Along the OISO-31 track, the difference between calculated and measured  $C_T$  is only 3.2  $\mu\text{mol kg}^{-1}$  (compared to the theoretical error of 7  $\mu\text{mol kg}^{-1}$  when using



**Fig. 2.** Relationship of  $A_T$  ( $\mu\text{mol kg}^{-1}$ ) versus Salinity deduced from surface  $A_T$  data obtained during OISO 11 and OISO-31 cruises in 2004 and 2021 (black circles) or from station data (at depth 0–20m from GLODAP data, grey triangles) in the South-Western Indian Ocean (zone 20–45°E/20–40°S). The relationship (grey line,  $A_T = 66.1773 * \text{Salinity} - 23.8222$ ) is used to calculate pH and  $C_T$  concentrations in this region.  $A_T$  underway data are available at NCEI/OCADS ([https://www.ncei.noaa.gov/access/ocean-carbon-data-system/oceans/VOS\\_Program/OISO.html](https://www.ncei.noaa.gov/access/ocean-carbon-data-system/oceans/VOS_Program/OISO.html)).

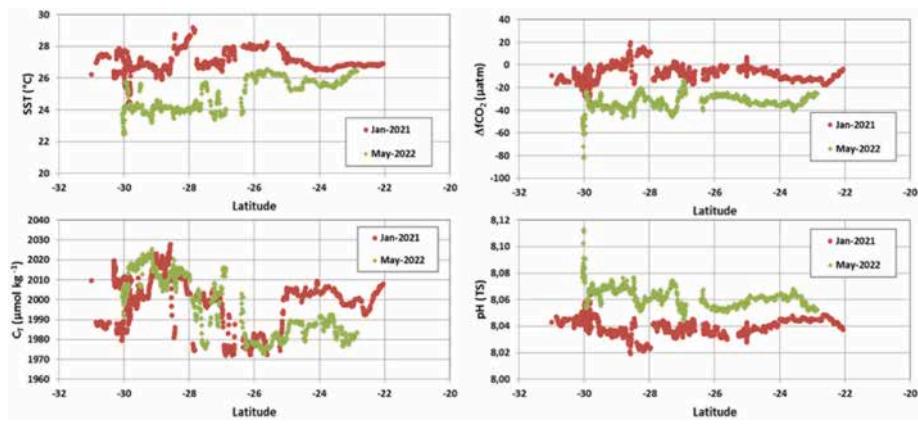
$f\text{CO}_2$  and  $A_T$  data). This supports the use of the selected  $A_T$ /S relationship for carbonate system calculations based on all  $f\text{CO}_2$  data available over 1963–2023 in the investigated region.

## 3. Results

### 3.1. Distribution during OISO-31 (January 2021) and RESILIENCE (May 2022)

The RESILIENCE cruise started from La Réunion in late April and went back to La Réunion in late May (Fig. 1). This cruise had two Legs: Leg 1 in the central Mozambique Channel (not included in this paper), and Leg 2 along the east coast of South Africa. The OISO-31 cruise also started from La Réunion in January 2021 and reached the African coast before moving at higher latitudes in the Southern Indian Ocean. Here we compare the data obtained along the same track between La Réunion and the African coast (Fig. 3). In January 2021, during the warm season,  $f\text{CO}_2$  was near equilibrium. In May 2022, the ocean was colder and acted as a  $\text{CO}_2$  sink for the atmosphere during this season ( $\Delta f\text{CO}_2 < 0$ , where  $\Delta f\text{CO}_2 = f\text{CO}_2^{\text{ocean}} - f\text{CO}_2^{\text{atm}}$ ). This is coherent with previous studies and  $\Delta f\text{CO}_2$  climatology (Poisson et al., 1993; Takahashi et al., 2009; Fay et al., 2024). In the subtropical Indian ocean north of 26°S,  $C_T$  and  $A_T$  concentrations were lower in May 2022 because the salinity was also lower and thus no difference was observed in salinity normalized  $C_T$  and  $A_T$ . Consequently the low  $f\text{CO}_2$  in May was mainly driven by temperature as generally observed in subtropics (e.g. Metzl et al., 1998; Sabine et al., 2000).

For both seasons the  $C_T$  concentrations were lower around 26°S south of Madagascar (1970  $\mu\text{mol kg}^{-1}$ ) when crossing a high productivity region (Fig. S2). The presence of low  $C_T$  concentrations was also observed around 40–43°E/28°S in 2021 and 2022 and were confirmed with direct  $C_T$  measurements in 2021 (Fig. S1). This was linked to the presence of a cyclonic eddy (as recorded in SST, Fig. 3) and a bloom occurring south-west of Madagascar (Fig. S2) associated to productive areas probably linked to  $\text{N}_2$ -fixers such as diazotrophs *Trichodesmium* (Poulton et al., 2009; Chowdhury et al., 2024). These local events changed slightly  $\Delta f\text{CO}_2$  (stronger sink) and  $C_T$ , but have no large impact when properties were averaged over the domain (Table 1). The highest



**Fig. 3.** Distribution of sea surface temperature,  $\Delta f\text{CO}_2$ ,  $C_T$ , and pH in January 2021 (red) and in May 2022 (green) along the same track in the southwestern Indian Ocean (from La Réunion to Durban). Average values are listed in Table 1.

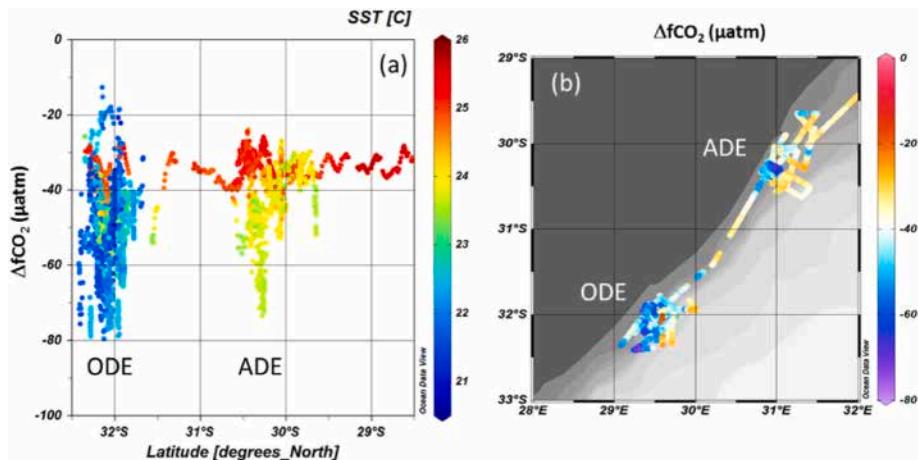
**Table 1**

Average of properties observed or calculated from  $f\text{CO}_2$  underway data in January 2021 (OISO-31) and in May 2022 (RESILIENCE) along the same track in the southwestern Indian Ocean (See Fig. 3). The mean values were estimated for the band 22–26°S, 26–31°S and 22–31°S. SD are in bracket. Nb is the number of data for each region. The difference (January minus May) are indicated.

Date	Region	SST °C	SSS psu	$f\text{CO}_2$ μatm	$\Delta f\text{CO}_2$ μatm	$A_T$ $\mu\text{mol.kg}^{-1}$	$C_T$ $\mu\text{mol.kg}^{-1}$	pH TS	$\Omega_{\text{Ca}}$	$\Omega_{\text{Ar}}$	Nb
Jan 2021	22–26°S	27.112 (0.429)	35.401 (0.122)	402.8 (4.8)	-7.4 (4.8)	2318.9 (8.1)	1998.9 (8.4)	8.039 (0.005)	5.47 (0.03)	3.63 (0.02)	436
Jan 2021	26–31°S	26.807 (0.739)	35.412 (0.172)	403.4 (7.9)	-6.8 (7.9)	2319.6 (11.4)	2002.3 (14.2)	8.039 (0.006)	5.43 (0.10)	3.60 (0.07)	1154
Jan 2021	22–31°S	26.890 (0.682)	35.409 (0.160)	403.2 (7.2)	-7.0 (7.2)	2319.5 (10.6)	2001.4 (12.9)	8.039 (0.006)	5.44 (0.09)	3.61 (0.07)	1590
May 2022	22–26°S	25.904 (0.379)	35.099 (0.091)	381.0 (3.8)	-31.6 (3.8)	2298.9 (6.0)	1982.9 (5.2)	8.058 (0.004)	5.39 (0.03)	3.56 (0.02)	279
May 2022	26–31°S	24.223 (0.647)	35.365 (0.192)	377.7 (7.2)	-34.9 (7.2)	2316.5 (12.7)	2008.1 (11.9)	8.065 (0.008)	5.24 (0.09)	3.45 (0.07)	599
May 2022	22–31°S	24.758 (0.972)	35.280 (0.208)	378.7 (6.5)	-33.8 (6.5)	2310.9 (13.7)	2000.1 (15.6)	8.062 (0.007)	5.29 (0.11)	3.49 (0.08)	878
Diff.	22–26°S	1.208	0.303	21.8	24.2	20.0	16.0	-0.018	0.08	0.07	
Diff.	26–31°S	2.583	0.047	25.7	28.1	3.1	-5.8	-0.025	0.18	0.15	
Diff.	22–31°S	2.133	0.129	24.5	26.9	8.5	1.3	-0.023	0.15	0.12	

calculated  $C_T$  concentrations ( $2020 \mu\text{mol.kg}^{-1}$ ) were observed around  $29^\circ\text{S}$  in 2021 and 2022 (Fig. 3). This is a location where the ocean with high temperature and high  $C_T$  was a  $\text{CO}_2$  source in January ( $\Delta f\text{CO}_2$  close to  $+20 \mu\text{atm}$ ). This is a local variation and we note that  $\Delta f\text{CO}_2$  was very homogeneous along this track in 2021 and 2022 (Fig. 3, Table 1), except near the coast where we observed a much stronger sink,  $\Delta f\text{CO}_2$  up to  $-80 \mu\text{atm}$  in May 2022.

During the RESILIENCE cruise (Leg 2), the coastal zone was investigated in detail at 2 locations: in an “Active Durban Eddy” (ADE, 4–7 May) located around  $30^\circ\text{S}$  and in an “Old Durban Eddy” (ODE 8–15 May) located at  $32^\circ\text{S}$  near the Protea Banks. The Durban Eddy at ADE was no longer active when the ship arrived on site while it has been clearly identified from satellite imagery a few days before. ADE are known to be regularly occurring temporary structures ( $\sim 10$  days) that



**Fig. 4.** Distribution of  $\Delta f\text{CO}_2$  ( $\mu\text{atm}$ ) during the RESILIENCE cruise in May 2022 along the African coast. In (a) color code is for SST ( $^\circ\text{C}$ ). In (b) color code is for  $\Delta f\text{CO}_2$  ( $\mu\text{atm}$ ). “ADE” and “ODE” identifies the locations of the Active Durban Eddy and the Old Durban Eddy investigated during the cruise. In May 2022, the ocean was always a  $\text{CO}_2$  sink in this region ( $\Delta f\text{CO}_2 < 0$ ). Figures produced with ODV (Schlitzer, 2018).

are then transported southwestwards through the Agulhas Current influence, to become ODE until they lose their kinetic energy and disappear (Guastella and Roberts, 2016). The distribution of  $\Delta f\text{CO}_2$  in May 2022 in this region is presented in Fig. 4. The ocean was always a  $\text{CO}_2$  sink ( $\Delta f\text{CO}_2 < -20 \mu\text{atm}$ ) as previously observed (Arnone et al., 2017). The lowest  $\Delta f\text{CO}_2$  ( $-60$  to  $-80 \mu\text{atm}$ ) were observed in the ADE and ODE rings. There was no clear link with the temperature or salinity and the low  $\Delta f\text{CO}_2$  observed at  $30$  and  $32^\circ\text{S}$  were driven by primary production. This is supported by the variation of surface Chl-a concentrations that present maxima occurring at  $29^\circ\text{S}$  and  $31$ – $30^\circ\text{S}$  in April 2022 (before the cruise) and in May 2022 at  $32^\circ\text{S}$  (Fig. S3). The observed  $\Delta f\text{CO}_2$  minima ( $<-50 \mu\text{atm}$  at  $29.5^\circ\text{S}$ ,  $30.5^\circ\text{S}$  and  $32^\circ\text{S}$ , Fig. 4) occurred where the sea surface Chl-a was high (up to  $3 \text{ mg m}^{-3}$ ). This supports that biological activity was the main driver of the  $f\text{CO}_2$  variations in May 2022 along the coast. This also suggests that biological ADE signature was still present at the time of the sampling while the eddy itself was no longer identifiable on site.

The contrasting  $f\text{CO}_2$  or  $\Delta f\text{CO}_2$  distribution near the coast and offshore described above was observed for most of cruises since 1963 (Fig. 5). The low  $\Delta f\text{CO}_2$  in April–May as observed in 2022 (up to  $-80 \mu\text{atm}$ ) was recorded only once (in late April 2018 at  $30^\circ\text{S}/31^\circ\text{E}$ ,  $\Delta f\text{CO}_2 = -50 \mu\text{atm}$ ). For most of the seasons, the high  $f\text{CO}_2$  variability occurred west of  $32^\circ\text{E}$ , i.e. west of the Agulhas current. The data also indicate a clear increase of  $f\text{CO}_2$  since 1963 in this region (Fig. 5). What is the decadal variability of  $f\text{CO}_2$  and pH in this region and is this signal driven by anthropogenic  $\text{CO}_2$  uptake or also due to natural variability? To explore the long-term trends of the carbonate system and given the spatial and seasonal variability of  $f\text{CO}_2$  in this region, we separated the domain in two regions, one in the Mozambique Basin east of  $32^\circ\text{E}$  (open ocean) and one near the coast (in the ADE and ODE regions).

### 3.2. Trends in the Mozambique basin

To evaluate the trend in the Mozambique Basin, we have selected the data in the region east of  $32^\circ\text{E}$  where  $f\text{CO}_2$  was homogeneous each year and season (Fig. 5). The mean values in this region since 1963 are presented in Fig. 6 and Table 2. The  $f\text{CO}_2$  seasonality being large of around

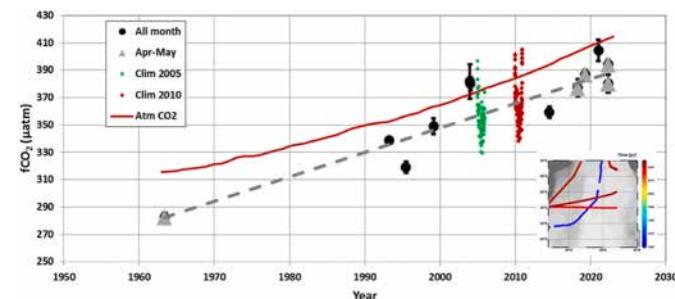
$40 \mu\text{atm}$  between summer and winter it is appropriate to select the same season for the trend analysis. We thus used the data available in April–May (only in 2018, 2019 and 2022), i.e. the same season as for the cruise conducted in 1963 (Keeling and Waterman, 1968). Over 1963–2022 the oceanic  $f\text{CO}_2$  trend of  $+1.79 \pm 0.13 \mu\text{atm.yr}^{-1}$  is close to the atmospheric trend ( $+1.66 \pm 0.03 \mu\text{atm.yr}^{-1}$ ). This explained that the  $\text{CO}_2$  sink was almost the same in May 1963 and May 2022 ( $\Delta f\text{CO}_2 = -33.1$  and  $-32.3 \mu\text{atm}$  respectively). This is also close to the climatology in May in this region ( $\Delta f\text{CO}_2 = -20.4 \pm 9.9$ , Fay et al., 2024).

As observed in the global ocean (Cheng et al., 2024) the South-West Indian Ocean experienced a long-term warming here evaluated at  $+0.120 \pm 0.009 \text{ }^\circ\text{C.decade}^{-1}$  since the 1960s (Fig. S4). As  $f\text{CO}_2$  is sensitive to temperature we corrected the  $f\text{CO}_2$  data to normalized temperature at  $25 \text{ }^\circ\text{C}$  following Takahashi et al. (1993):

$$f\text{CO}_2^{25} = f\text{CO}_2^{SST} * \text{EXP}(0.0423 * (25 - SST)) \quad (3)$$

The warming would have increased  $f\text{CO}_2$  by  $+9 \mu\text{atm}$  in 59 years, a signal included in the observations. When normalizing  $f\text{CO}_2$  at  $25 \text{ }^\circ\text{C}$ , the trend of  $f\text{CO}_2^{25}$  is  $+0.90 \pm 0.25 \mu\text{atm.yr}^{-1}$ , i.e. less than derived from  $f\text{CO}_2$ . To explain what drives the  $f\text{CO}_2$  (and pH) change, one must evaluate the anthropogenic  $\text{CO}_2$ .

To estimate the evolution of the anthropogenic  $\text{CO}_2$  concentrations ( $C_{\text{ant}}$ ) we selected the stations available from 1987 to 2021. The  $C_{\text{ant}}$  concentrations were calculated using the TrOCA method (Touratier et al., 2007) in the layer 100–150m as the back-calculation methods are not suitable for evaluating  $C_{\text{ant}}$  concentrations in surface waters due to gas exchange and biological activity. To estimate the  $C_{\text{ant}}$  trend, the concentrations were averaged for each period (Fig. 7a). As expected, the  $C_{\text{ant}}$  concentrations increase progressively from  $28.3 \pm 3.7 \mu\text{mol kg}^{-1}$  in 1987 to  $62.2 \pm 4.2 \mu\text{mol kg}^{-1}$  in 2021. The increase from 1995 to 2003 of  $+6.9 \mu\text{mol kg}^{-1}$  is in the range of the averaged  $C_{\text{ant}}$  increase of  $+7.7 \pm 0.5 \mu\text{mol kg}^{-1}$  observed between 1995 and 2003/2004 in the Indian Central Water (ICW) (Murata et al., 2010). Our results over 1987–2021 lead to a trend of  $C_{\text{ant}}$  of  $+1.03 \pm 0.14 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  (Table 3) and, as expected, the  $C_{\text{ant}}$  concentrations in the ocean are positively related to atmospheric  $\text{CO}_2$  (slope  $+0.512 \pm 0.050 \mu\text{mol kg}^{-1} \mu\text{atm}^{-1}$ , Fig. 7b). Interestingly the slope observed in the South-Western Indian Ocean over 1987–2021 is very close to that observed in the South Atlantic Central Water in the South Atlantic over 1972–2019 ( $+0.51 \pm 0.02 \mu\text{mol kg}^{-1} \mu\text{atm}^{-1}$ , Fontela et al., 2021). Furthermore, the trend of  $C_{\text{ant}}$  in the Mozambique Basin is also comparable to that derived further east at  $30^\circ\text{S}$ – $55^\circ\text{E}$  in the Madagascar Basin between 1978 and 2020 ( $+1.05 \pm 0.08 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$ , Metzl et al., 2022) indicating this signal is homogeneous in the southern Atlantic and Indian oceans at these latitudes.



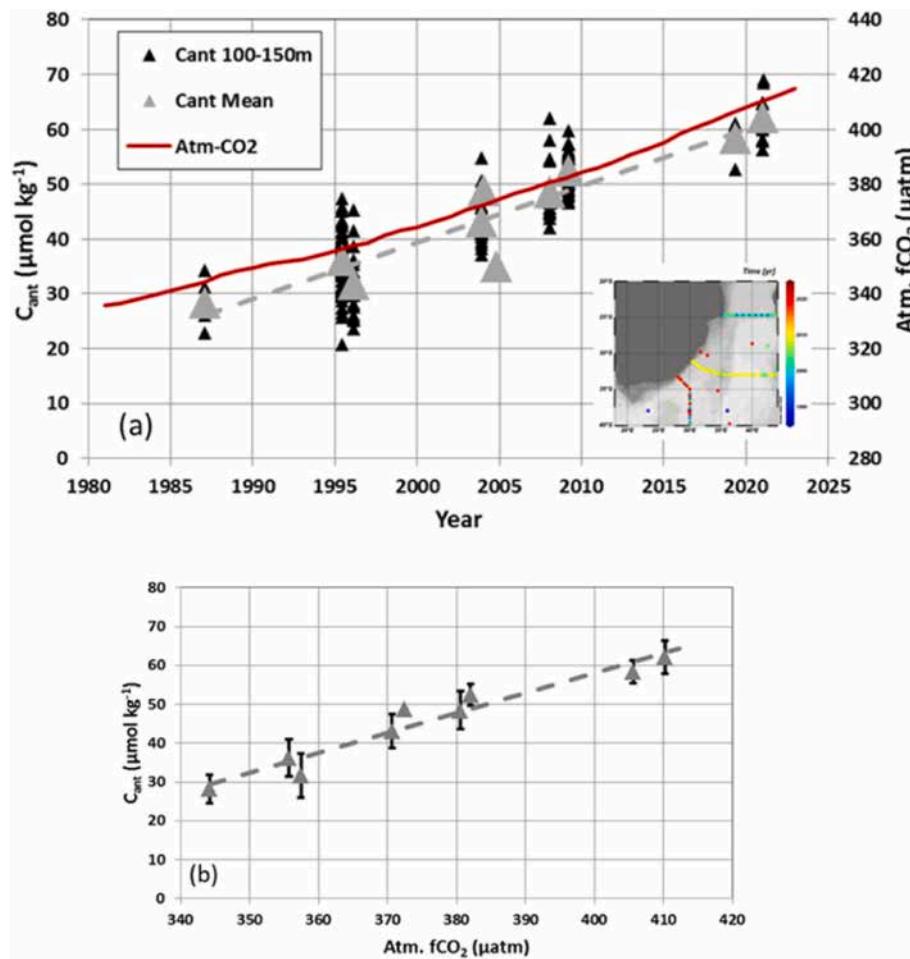
**Fig. 6.** Time-series of monthly mean  $f\text{CO}_2$  ( $\mu\text{atm}$ , black dots for all season and grey triangles for April–May) in the Mozambique Basin (region  $24^\circ\text{S}$ – $35^\circ\text{S}$ / $32^\circ\text{E}$ – $42^\circ\text{E}$ ). The red curve is the atmospheric  $f\text{CO}_2$ . The monthly climatology for a reference year 2005 or 2010 (Takahashi et al., 2014; Fay et al., 2024) is also plotted (green and red diamonds) highlighting the seasonality in this region. For the period April–May the oceanic  $f\text{CO}_2$  increase of  $+1.79 \pm 0.13 \mu\text{atm.yr}^{-1}$  (dashed grey line) is close to the atmospheric increase. The selected tracks in April–May are indicated in the insert map produced with ODV (Schlitzer, 2018). The mean monthly values and standard deviations are listed in Table 2.

**Fig. 5.** Distribution of sea surface  $f\text{CO}_2$  ( $\mu\text{atm}$ ) in the eastern African coastal zone and offshore for all cruises over 1963–2023 in the SOCAT data-base, v2024 (Bakker et al., 2016, 2024); color code is for Year. This includes the OISO-31 and RESILIENCE cruises. The selected tracks are indicated in the insert map. Figures produced with ODV (Schlitzer, 2018).

**Table 2**

Average of properties measured or calculated from the sea surface underway  $f\text{CO}_2$  data in the Mozambique Basin. The mean values were estimated for the box 24°box24°S–35°S/32°E–42°E (insert map in Fig. 6). SD are in bracket. Nb is the number of data for each region. Bold value identifies anomalies in 2019.

Year	Month	NB	SST	SSS	$f\text{CO}_2$	$A_T$	$C_T$	pH	$\Omega_{\text{Ca}}$	$\Omega_{\text{Ar}}$
1963	5	54	22.133 (1.027)	35.369 (0.156)	282.3 (3.5)	2316.8 (10.3)	1963.8 (13.1)	8.168 (0.006)	5.90 (0.11)	3.86 (0.08)
1993	3	10	25.486 (0.034)	35.437 (0.016)	338.6 (1.9)	2321.3 (1.0)	1977.0 (1.7)	8.102 (0.002)	5.82 (0.01)	3.84 (0.01)
1995	6	2876	23.290 (0.702)	35.489 (0.053)	319.1 (4.2)	2324.7 (3.5)	1985.9 (7.0)	8.125 (0.005)	5.70 (0.09)	3.74 (0.06)
1999	2	403 (0.06)	26.333 (0.106)	35.388 (5.7)	348.9 (7.0)	2318.1 (10.4)	1973.8 (0.005)	8.090 (0.08)	5.84 (0.06)	3.86 (0.524)
2003	12	450	27.137 (1.006)	35.297 (0.067)	381.9 (12.5)	2312.1 (4.4)	1982.1 (6.5)	8.057 (0.012)	5.63 (0.07)	3.74 (0.06)
2004	1	195	27.159 (0.413)	35.344 (0.057)	380.1 (4.8)	2315.1 (3.8)	1983.2 (5.5)	8.059 (0.005)	5.66 (0.05)	3.76 (0.04)
2014	7	882 (0.06)	23.506 (0.054)	35.342 (4.0)	359.3 (3.6)	2315.0 (7.0)	2002.6 (0.004)	8.083 (0.08)	5.29 (0.06)	3.48 (0.683)
2018	4	945	25.126 (1.320)	35.500 (0.178)	377.1 (6.4)	2325.5 (11.8)	2006.5 (15.2)	8.065 (0.007)	5.42 (0.14)	3.58 (0.11)
2019	4	120	<b>27.290</b> (0.159)	<b>35.328</b> (0.095)	386.8 (3.5)	<b>2314.1</b> (6.3)	<b>1985.1</b> (4.6)	8.053 (0.004)	5.62 (0.03)	3.73 (0.02)
2021	1	757	26.676 (0.588)	35.509 (0.118)	404.6 (7.9)	2326.1 (7.8)	2008.7 (11.1)	8.039 (0.006)	5.42 (0.08)	3.60 (0.06)
2022	4	173	26.407 (0.400)	35.412 (0.039)	394.2 (3.2)	2319.6 (2.6)	2000.8 (3.2)	8.048 (0.003)	5.44 (0.08)	3.60 (0.06)
2022	5	720	24.912 (1.039)	35.459 (0.084)	380.3 (6.6)	2322.8 (5.6)	2008.2 (7.8)	8.062 (0.007)	5.35 (0.13)	3.53 (0.09)



**Fig. 7.** (a) Time-series of anthropogenic CO<sub>2</sub> concentrations (C<sub>ant</sub>) over 1987–2021 estimated in subsurface (layer 100–150m) from the GLODAP-v2023 data (Lauvset et al., 2024) completed with OISO cruise in 2021 (location of selected stations in the insert map, color code is for year). The figure shows the C<sub>ant</sub> concentrations calculated for each sample (black) and the C<sub>ant</sub> averaged in the layer 100–150m for each period (grey triangles). Over the period 1987–2021, the C<sub>ant</sub> trend is  $+1.03 \pm 0.14 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  (dashed grey line). The red curve is the atmospheric  $f\text{CO}_2$ . (b): same data for C<sub>ant</sub> versus atmospheric  $f\text{CO}_2$  (slope =  $+0.512 \pm 0.050 \mu\text{mol kg}^{-1} \mu\text{atm}^{-1}$ ).

**Table 3**

Trends of  $C_{\text{ant}}$  ( $\mu\text{mol} \cdot \text{kg}^{-1} \cdot \text{yr}^{-1}$ ) in the Mozambique basin and the coastal zone in the layer 100–150 m for different periods. Standard-deviations are given in brackets.

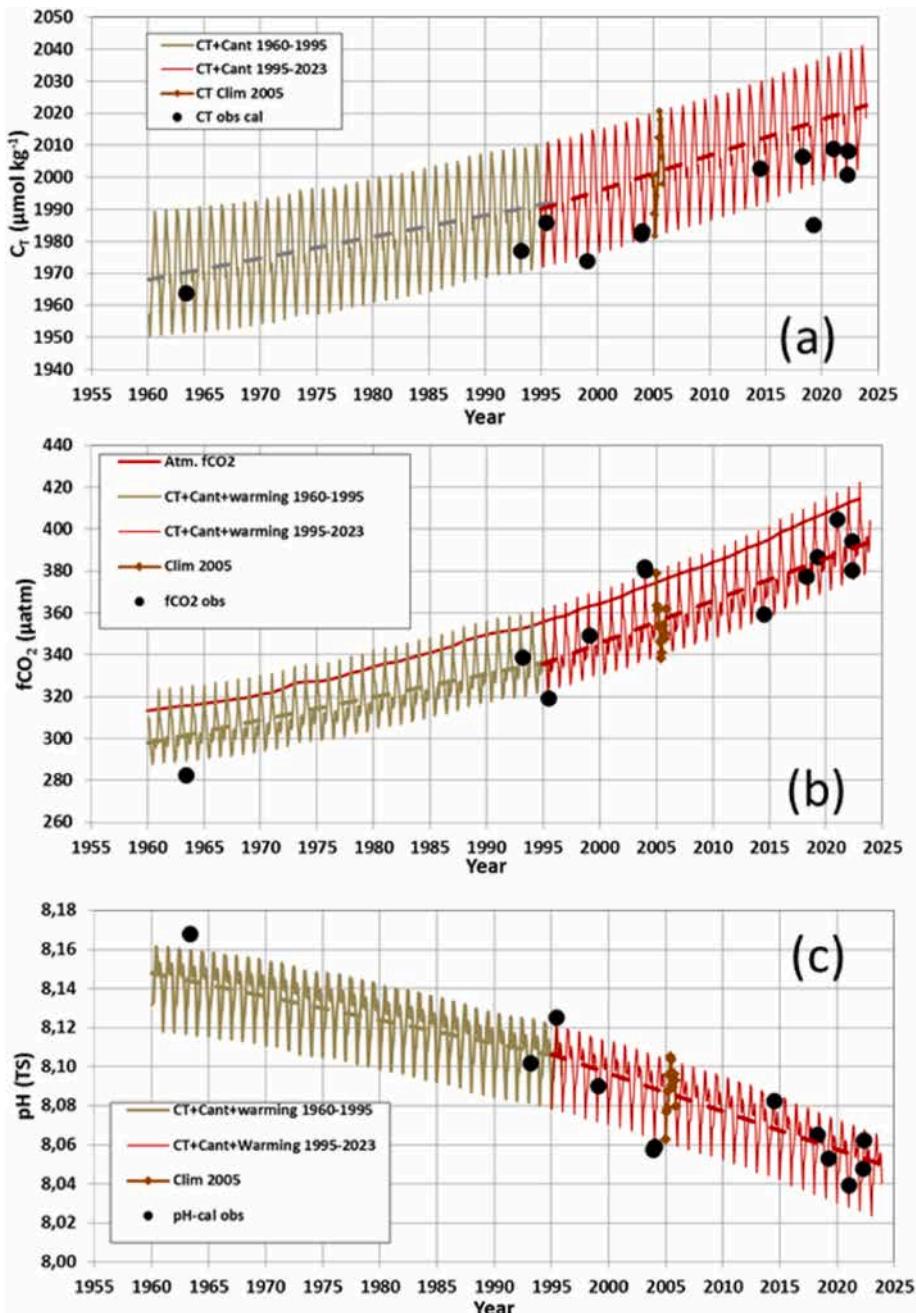
Zone	Period	Trend $C_{\text{ant}}$ $\mu\text{mol} \cdot \text{kg}^{-1} \cdot \text{yr}^{-1}$
Mozambique Basin	1987–2021	+1.03 (0.14)
Mozambique Basin	1995–2021	+1.02 (0.11)
Coastal zone	1995–2021	+0.74 (0.21)

At constant temperature and alkalinity, the  $C_{\text{ant}}$  trend would lead to an increasing rate for  $f\text{CO}_2$  of  $+1.58 \mu\text{atm} \cdot \text{yr}^{-1}$  slightly lower than observed in surface waters (Fig. 6). For pH this would translate in the decrease of  $-0.017 \cdot \text{decade}^{-1}$ .

To interpret the observed  $f\text{CO}_2$  change since the 1960s (Fig. 6) we assumed that the  $C_{\text{ant}}$  trend evaluated in subsurface represents the  $C_{\text{ant}}$  change in surface and we calculated the monthly  $C_T$  concentration over 1960–2022 by correcting  $C_T$  for each year using the relationship observed between  $C_{\text{ant}}$  and atmospheric  $\text{CO}_2$  (Fig. 7b).

$$C_T(t) + C_T(t-1) + C_{\text{ant}}(t) - C_{\text{ant}}(t-1) \quad (4)$$

In this scenario we assumed that the  $C_T$  seasonality is not changing over



**Fig. 8.** Observed (black dots) and calculated time-series in the Mozambique Basin for (a)  $C_T$  ( $\mu\text{mol} \cdot \text{kg}^{-1}$ ), (b)  $f\text{CO}_2$  ( $\mu\text{atm}$ ) and (c) pH (TS). The lines show the evolution of the properties since 1960 corrected for  $C_{\text{ant}}$  where  $f\text{CO}_2$ , pH were recalculated using reconstructed  $C_T$ . Also shown the climatology for year 2005 (red diamonds, [Takahashi et al., 2014](#)). In (b) the red line is the atmospheric  $f\text{CO}_2$ . The trends (dashed line) are shown over 1960–1995 (brown) and 1995–2023 (red) (trends values are listed in Table 4).

time (Fig. 8a). The  $f\text{CO}_2$  and pH are calculated using the reconstructed monthly  $C_T$  concentrations and the seasonal  $A_T$  based on the climatology (Takahashi et al., 2014). We also take into account the warming of  $+0.12^\circ\text{C}.\text{decade}^{-1}$  for  $f\text{CO}_2$  and pH calculations (Fig. 8b and c). The reconstructed time series of  $C_T$  confirms that the  $C_{\text{ant}}$  accumulation explained most of the temporal  $C_T$  change observed in surface waters between 1963 and 2022.

The simulated  $C_T$  is in the range of observations except in April 2019 where the low  $C_T$  calculated from  $f\text{CO}_2$  data was associated to salinity and temperature anomalies (Table 2). For other periods the mean difference of  $C_T$  between the reconstructed time-series and the observations is  $+6.6 \pm 8.3$  ( $n = 10$ )  $\mu\text{mol kg}^{-1}$ . Consequently, the results indicate that the increase of oceanic  $f\text{CO}_2$  and the decrease of pH are driven by the anthropogenic  $\text{CO}_2$  uptake and the ocean warming, the latter representing about 10% of the change in the recent decades (Table 4). The lowest pH of 8.04 was observed in January 2021. In May 2022 the pH was 8.062, i.e. -0.106 lower than in May 1963 (Fig. 8c) a difference equal to the mean change of -0.11 in the global ocean since the pre-industrial era (Jiang et al., 2019). The reconstructed time-series also shown that the trends are different depending the decade. The trends of  $C_T$ ,  $f\text{CO}_2$  and pH over the last three decades (1995–2023) were faster than over 1960–1995 (Table 4). These results are coherent with those investigated in the southern Mozambique Channel (Lo Monaco et al., 2021). In the Mozambique Basin where  $f\text{CO}_2$  is rather homogeneous from time to time (Fig. 5), upwelling or biological activity have small impact on the  $C_T$  or the  $A_T$  variability, excepted when large bloom occurred south of Madagascar (Metzl et al., 2022) as also observed in 2021 and 2022 around 28°S (Fig. 3).

We now attempt to explore the  $f\text{CO}_2$  and pH trends in the coastal zone where the interpretation is more obscure as the anthropogenic  $\text{CO}_2$  and the warming effect on  $f\text{CO}_2$  trends are probably masked by the variability in circulation near the Agulhas Current, the coastal upwelling or the biological activity as depicted from Chl-a (Fig. S3).

### 3.3. Trends in the coastal zone

As described above, the variability of  $f\text{CO}_2$  in the Agulhas frontal system and near the coast was very large in May 2022 (Fig. 4). To explore the trends in this region we selected two regions, the ADE around 30°S and the ODE around 32°S that were visited during the RESILIENCE cruise. In the region 30–32°S/30–32°E there are few historical stations, available only in June 1995, March 2009 and January 2021. Over this period we estimated a  $C_{\text{ant}}$  trend of  $+0.74 \pm 0.21 \mu\text{mol}$

$\text{kg}^{-1} \text{yr}^{-1}$  in the layer 100–150m (Fig. S5) smaller than evaluated in the Mozambique Basin ( $+1.020 \pm 0.106 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  when using the same period, 1995–2021, Table 3). The relationship with atmospheric  $\text{CO}_2$  is also different from the one deduced in the Mozambique Basin ( $C_{\text{ant}} = +0.342 \pm 0.119 \mu\text{mol kg}^{-1} \mu\text{atm}^{-1}$ ). Why the  $C_{\text{ant}}$  trend was lower in the coastal zone is not clear and for the reconstruction of the carbonate properties, we tested both relations. Although the  $C_{\text{ant}}$  trends in the Mozambique Basin and in the coastal zone are different, the  $C_T$  reconstructions seems almost the same (Fig. S6) but the comparison with observations suggested the reconstruction using  $C_{\text{ant}}$  trend in the coastal zone was better for this region (Fig. 9). The reconstructed  $C_T$  in the coastal region is in the range of the observations except in 2008 (at 32°S) and in 2021 (at 30°S) where the low  $C_T$  were linked to a warm anomaly. In May 2022 the concentration of reconstructed  $C_T$  (2014  $\mu\text{mol kg}^{-1}$ ) was almost the same as deduced from the  $f\text{CO}_2$  data (2000  $\mu\text{mol kg}^{-1}$  at 30°S in the ADE and 2012  $\mu\text{mol kg}^{-1}$  at 32°S in the ODE). The recalculated  $f\text{CO}_2$  was also very close to the observations in 2022 and 2023 (Fig. 9b). In June 1995 at 30°S we noticed lower observed  $f\text{CO}_2$  ( $-10 \mu\text{atm}$  compared to the reconstruction) that might be linked to biological event but this cannot be confirmed with Chl-a data in 1995.

The trend of  $C_T$  in the coastal region was faster in the recent decade (1995–2023) but lower compared to the trend evaluated in the Mozambique Basin (Tables 4 and 5). Consequently, the annual rate of oceanic  $f\text{CO}_2$  was lower in the coastal region ( $1.37 \pm 0.07 \mu\text{atm yr}^{-1}$ ) (Table 5) resulting in an increasing ocean  $\text{CO}_2$  sink over 1995–2023 ( $\Delta f\text{CO}_2$  trend  $= -0.75 \pm 0.07 \mu\text{atm yr}^{-1}$ ). This contrasts with the results in the Mozambique Basin where the  $\text{CO}_2$  sink was stable over 1995–2023 ( $\Delta f\text{CO}_2$  trend  $= -0.09 \pm 0.07 \mu\text{atm yr}^{-1}$ , Fig. S7). In the coastal zone, we found that the ocean was near equilibrium ( $-5 < \Delta f\text{CO}_2 < 0 \mu\text{atm}$ ) in the 1960s and moved to a sink of  $\text{CO}_2$  during the last 2 decades ( $\Delta f\text{CO}_2 < -20 \mu\text{atm}$ ). The lowest  $\Delta f\text{CO}_2$  ( $-80 \mu\text{atm}$ ) was observed in May 2022 in both the ADE and ODE regions (Fig. 4). The lowest pH (8.045) in the coastal zone was observed at 30°S in January 2021 (Fig. 9c). In May 2022 the pH of 8.071 was higher by 0.026, confirming the seasonal difference (May–January) of 0.03 in the climatology (Takahashi et al., 2014). Since 1995, the pH trend in the coastal region appeared slower ( $-0.014 \pm 0.001 \cdot \text{decade}^{-1}$ ) than in the Mozambique Basin ( $-0.020 \pm 0.001 \cdot \text{decade}^{-1}$ ). Our results in the eastern African coast is about half the pH trend of  $-0.03 \pm 0.01 \cdot \text{decade}^{-1}$  estimated in the western coastal side but over a short period 2005–2012 (González-Dávila et al., 2017).

### 3.4. Air-sea $\text{CO}_2$ fluxes

The increasing of the  $\text{CO}_2$  sink deduced from our results in the southern African coastal zone (here the MARCATS #25) is in line with previous data-based analysis in other shelves regions (Laruelle et al., 2018). This is also coherent with models results over 1998–2018 in this region (Resplandy et al., 2024; Mathis et al., 2024). Our recent observations in 2021–2022 also showed that the ocean  $\text{CO}_2$  sink continued to increase (Fig. S7) that might be explained by lateral transport as suggested from modeling study (Roobaert et al., 2024).

The net flux of  $\text{CO}_2$  across the air-sea interface ( $FCO_2$ ) was calculated according to the following equation (5):

$$FCO_2 = k K_0 \Delta f\text{CO}_2 \quad (5)$$

Where  $K_0$  is the solubility of  $\text{CO}_2$  in seawater calculated from in situ temperature and salinity (Weiss, 1974) and  $k$  ( $\text{cm.h}^{-1}$ ) is the gas transfer velocity expressed from the wind speed  $U$  ( $\text{m.s}^{-1}$ ) (Wanninkhof, 2014) and the Schmidt number  $Sc$  (Wanninkhof, 1992) following equation (6):

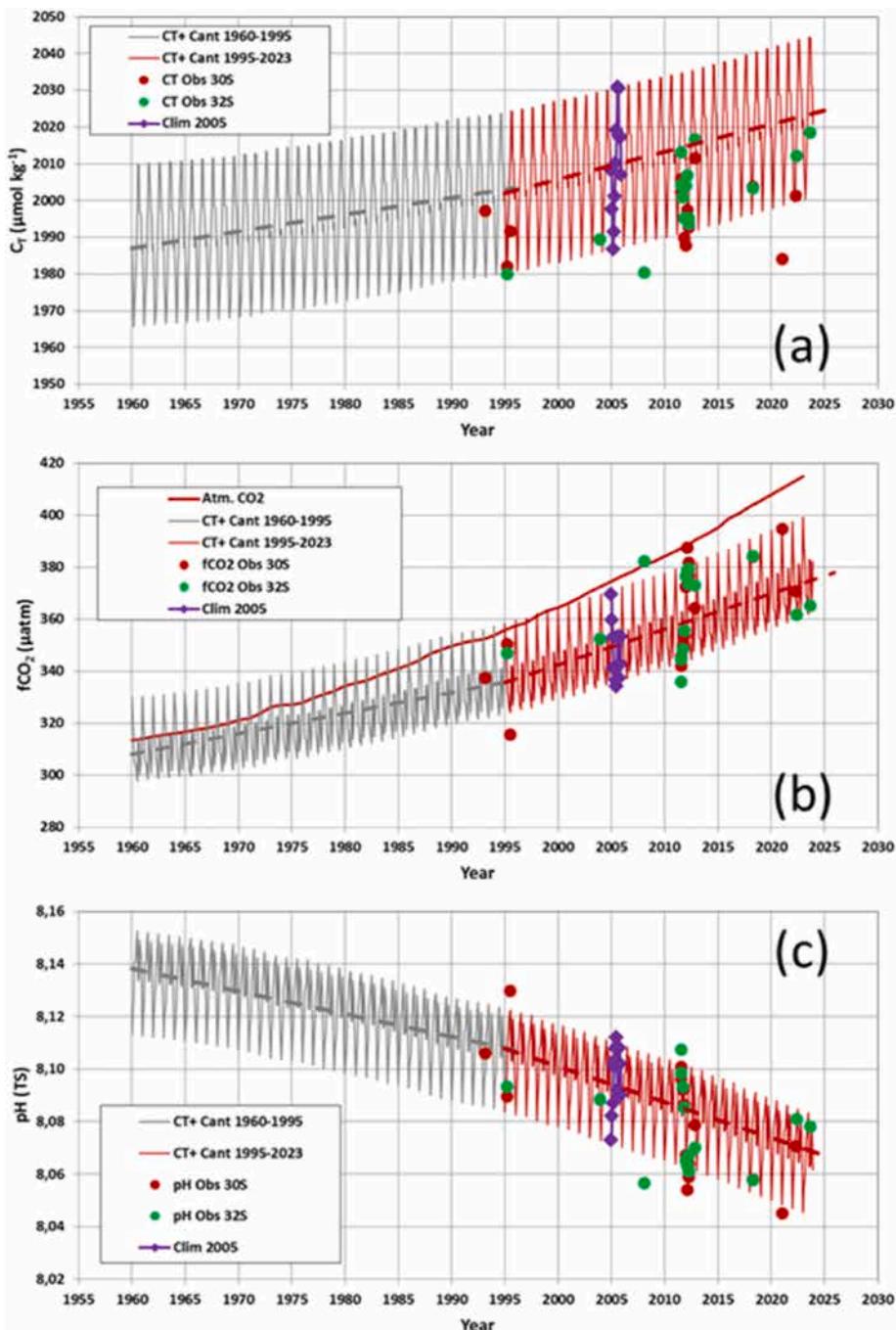
$$k = 0.251 U^2 (Sc/660)^{-0.5} \quad (6)$$

For the wind speed, we used the monthly values from MERRA-2 for 1980–2023 and the mean monthly values for years 1960–1979. In the coastal zone, winds are slightly higher than in the open ocean and in

**Table 4**

Trends of sea surface  $C_T$  ( $\mu\text{mol} \cdot \text{kg}^{-1} \cdot \text{yr}^{-1}$ ),  $f\text{CO}_2$  ( $\mu\text{atm} \cdot \text{yr}^{-1}$ ) and pH (TS.  $\text{decade}^{-1}$ ) based on observations (Obs.) and reconstruction (Rec.) for different periods in the Mozambique Basin (Fig. 8). Standard-deviations are given in brackets. Results for the reconstruction without warming are also listed (no warming).

Zone	Season	Trend $C_T$ $\mu\text{mol} \cdot \text{kg}^{-1} \cdot \text{yr}^{-1}$	Trend $f\text{CO}_2$ $\mu\text{atm} \cdot \text{yr}^{-1}$	Trend pH TS. $\text{decade}^{-1}$	Method
1963–2022	Apr–May	0.64 (0.20)	1.78 (0.13)	-0.019 (0.001)	Obs.
1960–1995	Annual	0.67 (0.05)	1.09 (0.05)	-0.012 (0.001)	Rec.
1995–2023	Annual	1.11 (0.08)	2.03 (0.07)	-0.020 (0.001)	Rec.
1960–1995	Annual	0.67 (0.05)	0.94 (0.05)	-0.010 (0.001)	no warming
1995–2023	Annual	1.11 (0.08)	1.84 (0.07)	-0.018 (0.001)	no warming
Warming effect	1960–1995		13.8 %	16.7 %	
Warming effect	1995–2023		9.4 %	10.0 %	



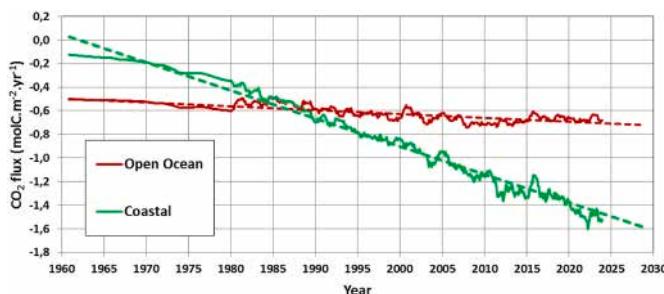
**Fig. 9.** Observed (red dots at 30°S for ADE, green dots at 32°S for ODE) and calculated time-series in the coastal region off Africa for (a)  $C_T$  ( $\mu\text{mol} \cdot \text{kg}^{-1}$ ), (b)  $f\text{CO}_2$  ( $\mu\text{atm}$ ) and (c) pH (TS). The lines show the evolution of the properties since 1960 corrected for  $C_{\text{ant}}$  where  $f\text{CO}_2$ , pH were recalculated using reconstructed  $C_T$ . Also shown the climatology for year 2005 (Purple diamonds, [Takahashi et al., 2014](#)). In (b) the red line is the atmospheric  $f\text{CO}_2$ . The trends (dashed line) are shown over 1960–1995 (grey) and 1995–2023 (red) (trends values are listed in [Table 5](#)).

**Table 5**

Trends of sea surface  $C_T$  ( $\mu\text{mol} \cdot \text{kg}^{-1} \cdot \text{yr}^{-1}$ ),  $f\text{CO}_2$  ( $\mu\text{atm} \cdot \text{yr}^{-1}$ ) and pH (TS.  $\text{decade}^{-1}$ ) based on reconstruction method (Rec.) for different periods in the coastal region. See [Fig. 9](#). Standard-deviations are given in brackets.

Period	Season	Trend $C_T$ $\mu\text{mol} \cdot \text{kg}^{-1} \cdot \text{yr}^{-1}$	Trend $f\text{CO}_2$ $\mu\text{atm} \cdot \text{yr}^{-1}$	Trend pH TS. $\text{decade}^{-1}$	Method
1960–1995	Annual	0.45 (0.06)	0.79 (0.04)	-0.0087 (0.0005)	Rec.
1995–2023	Annual	0.75 (0.09)	1.37 (0.07)	-0.014 (0.001)	Rec.

both regions no clear trend was detected for the wind since 1980 ([Fig. S8](#)). The long-term change of the  $\text{CO}_2$  sink ([Fig. 10](#)) was mainly driven by  $\Delta f\text{CO}_2$  ([Fig. S7](#)). Inter-annual variability of the sink is observed during some periods such as in 2015 when the wind was relatively low in both regions but this has no effect on the trends. The trends of fluxes over 1960–2023 were  $-0.0238 \pm 0.0001 \text{ molC/m}^2/\text{yr}$  in the coastal zone and only  $-0.0058 \pm 9.2 \text{ molC/m}^2/\text{yr}$  in the Mozambique basin ([Fig. 10](#)). In the Mozambique basin the sink was  $-0.50 \text{ molC/m}^2/\text{yr}$  in 1960 and increased to  $-0.77 \text{ molC/m}^2/\text{yr}$  in 2022. In the coastal zone the sink was  $-0.13 \text{ molC/m}^2/\text{yr}$  in 1960 and increased to  $-1.52 \text{ molC/m}^2/\text{yr}$  in 2022 ([Table 6](#)). Over 2005–2012 we estimated an average sink



**Fig. 10.** Reconstructed time-series of the  $\text{CO}_2$  fluxes ( $\text{molC.m}^{-2}.\text{yr}^{-1}$ ) in the Mozambique Basin (open ocean region, red) and in the African coastal zone (green). Corresponding linear trends are indicated in dashed.

of  $-1.16 \pm 0.08 \text{ molC.m}^{-2}.\text{yr}^{-1}$  in the coastal zone that is 30% lower than  $-1.65 \pm 0.04 \text{ molC.m}^{-2}.\text{yr}^{-1}$  previously reported by Arnone et al. (2017) based on  $\text{fCO}_2$  observations for the same period (Table 6). Compared to ocean models (Bourgeois et al., 2016), our reconstructed results in the Agulhas coastal zone (region MARCATS #25) for 1990–2011 ( $-0.93 \pm 0.17 \text{ molC.m}^{-2}.\text{yr}^{-1}$ ) are in the same range but stronger than deduced from data-based product for the same period (Laruelle et al., 2014, Table 6). The  $\text{CO}_2$  flux deduced from observations in 1963–2023 showed a clear seasonality (Fig. S9a) but there are not enough observations each year to integrate annual values (Fig. S9b). This is why the fluxes deduced from observations are presented in  $\text{mmolC.m}^{-2}.\text{d}^{-1}$  (Table 6). However, they confirmed that both regions were acting as an ocean  $\text{CO}_2$  sink except in the Mozambique basin in December 2003/January 2004 (Table 6, Fig. S9b) when the region was warmer (Fig. S4).

#### 4. Summary and concluding remarks

The observations obtained in January 2021 and May 2022 presented in this study offered a new description of the spatial and temporal variability of the carbonate system in the South Western Indian Ocean,

the Agulhas Current system and near the African coast. For the seasonal cycle, the new data in May 2022 complemented previous cruises conducted in January–April and June–December (Bakker et al., 2016, 2024). In the Mozambique Basin the  $\text{fCO}_2$  distribution was rather homogeneous in January and May with some lower  $\text{fCO}_2$  and  $\text{C}_T$  concentrations localized south of Madagascar associated to productive areas probably linked to diazotrophy. In this region the ocean was a small  $\text{CO}_2$  sink in January 2021 ( $-0.5 \text{ mmolC.m}^{-2}.\text{d}^{-1}$ ) and much stronger in May 2022 during a colder season ( $-2.4 \text{ mmolC.m}^{-2}.\text{d}^{-1}$ ). In the Agulhas Current system and near the African coast the variability of  $\text{fCO}_2$  was very large with  $\Delta\text{fCO}_2$  ranging between  $-20$  and  $-80 \mu\text{atm}$  in May 2022. Based on observations we estimated that the coastal zone was a strong sink in May 2022 ( $-4.3 \text{ mmolC.m}^{-2}.\text{d}^{-1}$ ). To explore the trends and the drivers of the ocean carbonate system at decadal scale, we separated the domain following two regions, the Mozambique Basin and the coastal areas. Using sea surface  $\text{fCO}_2$  data available since 1995 we found a faster  $\text{fCO}_2$  increase in the Mozambique Basin ( $2.03 \pm 0.07 \mu\text{atm.yr}^{-1}$ ) compared to the coastal zone ( $1.37 \pm 0.07 \mu\text{atm.yr}^{-1}$ ). From the water column data obtained between 1987 and 2021, we estimated positive trends of the anthropogenic  $\text{CO}_2$  concentrations in subsurface that enabled to reconstruct the temporal changes of the carbonate system since 1960. The reconstructed air-sea  $\text{CO}_2$  fluxes indicate that the oceanic  $\text{CO}_2$  sink increased over time, a change that was more pronounced in the coastal zone. The accumulation of anthropogenic  $\text{CO}_2$  drove the continuous increase of  $\text{fCO}_2$  and  $\text{C}_T$  and the decrease of pH. In the Mozambique Basin, the lowest pH (8.039) was observed in January 2021. It was higher in May 2022 (8.062) and lower by  $-0.106$  than observed 59 years ago for the same season (May 1963); this is close to the difference of  $-0.11$  estimated on average for the global ocean since the pre-industrial era (Jiang et al., 2019). In the coastal zone the lowest pH (8.045) was also observed in January 2021 and highly variable in May 2022 with values ranging between 8.05 and 8.11. In both regions, the pH decrease was about twice faster in the last 3 decades (1995–2023). The pH trend was faster in the open ocean ( $-0.020 \pm 0.001.\text{decade}^{-1}$ ) compared to the coastal zones ( $-0.014 \pm 0.001$ ).

**Table 6**

Air-sea  $\text{CO}_2$  fluxes estimated in the investigated region from different methods and for different periods (negative values represent an ocean  $\text{CO}_2$  sink). Annual fluxes are expressed in  $\text{molC.m}^{-2}.\text{yr}^{-1}$ ; observational fluxes for the OISO-31 cruise in January 2021 and RESILIENCE cruise in May 2022 expressed in  $\text{molC.m}^{-2}.\text{d}^{-1}$ . Standard-deviations are given in brackets. For the methods, the results based on reconstruction noted Rec. and from observations noted Obs.

Region		Flux $\text{molC.m}^{-2}.\text{yr}^{-1}$	Method	Year/Period	Ref
Open Ocean	37.5°E–32°S	-0.66	Climatology	2000	Takahashi et al. (2009)
Open Ocean	37.5°E–32°S	-1.04	Climatology	2010	Fay et al. (2024)
Open Ocean	32.5°E–32°S	-1.13	Climatology	2000	Takahashi et al. (2009)
Open Ocean	32.5°E–32°S	-1.70	Climatology	2010	Fay et al. (2024)
Coastal Zone	30°E–32°S	-1.65 (0.04)	Observations	2005–2012	Arnone et al. (2017)
Coastal Zone	MARCATS #25	-0.58	Data-Based	1990–2011	Laruelle et al. (2014)
Coastal Zone	MARCATS #25	-1.20 (0.09)	Model	1990–2011	Bourgeois et al. (2016)
Open Ocean	Mozambique	-0.74	Rec.	2000	This study
Open Ocean	Mozambique	-0.80	Rec.	2010	This study
Coastal Zone	30°S	-1.16	Rec.	2010	This study
Coastal Zone	30°S	-1.16 (0.08)	Rec.	2005–2012	This study
Coastal Zone	30°S	-0.93 (0.17)	Rec.	1990–2011	This study
Open Ocean	Mozambique	-0.50	Rec.	1960	This study
Open Ocean	Mozambique	-0.77	Rec.	2022	This study
Coastal Zone	30°S	-0.13	Rec.	1960	This study
Coastal Zone	30°S	-1.52	Rec.	2022	This study
Region		Flux $\text{mmolC.m}^{-2}.\text{d}^{-1}$	Method	Year/Period	Ref
Open Ocean	Mozambique	-2.39	Rec.	May 2022	This study
Coastal Zone	30°S	-3.38	Rec.	May 2022	This study
Open Ocean	Mozambique	+0.87	Obs.	Jan 2004	This study
Open Ocean	Mozambique	-0.46	Obs.	Jan 2021	This study
Open Ocean	Mozambique	-2.37	Obs.	May 2022	This study
Coastal Zone	30°S	-3.51	Obs.	May 2022	This study
Coastal Zone	32°S	-4.30	Obs.	May 2022	This study

decade<sup>-1</sup>). The rate of OA deduced from our reconstruction is low compared to the first estimate in the Indian Ocean Subtropics for the period 1991–2011 ( $-0.027 \pm 0.01$ .decade<sup>-1</sup>, Lauvset et al., 2015) but in the range of recent neural network data-based method ( $-0.018 \pm 0.001$ .decade<sup>-1</sup>, Chau et al., 2024). Although there are relatively few data available at different seasons and years, our recent observations confirmed the acidification deduced from the reconstruction (Figs. 8 and 9). In austral summer the observed pH in the coastal zone changed from 8.106 in February 1993 to 8.045 in January 2021. In austral autumn, observed pH changed from 8.130 in June 1995 to 8.070 May 2022. The decrease of pH was mainly driven by anthropogenic CO<sub>2</sub> uptake and also linked to the long term warming that represents about 10% of the pH changes.

Given the high variability of the carbonate system in the Agulhas Current system and in the coastal zone (here the MARCATS #25 region) more observations and dedicated models are needed to better evaluate and understand the processes that control the air-sea CO<sub>2</sub> fluxes and ocean acidification in such dynamics region, as well as the impact of chemistry change on the marine ecosystem in the future. Our present study based on fCO<sub>2</sub>, A<sub>T</sub> and C<sub>T</sub> data calls for detailed analyses that couple chemical and biological observations, such as dinitrogen fixation that present high rates near the South African coast and in the South Western Indian Ocean (Chowdhury et al., 2024). Our analysis was dedicated to evaluate changes of the carbonate system in surface waters. In subsurface, we estimated C<sub>ant</sub> concentrations close to 60 μmol kg<sup>-1</sup> in 2021, a signal identified down to 500m. This should be investigated with new observations in the future as it has been recently recognized that this region present significant decadal differences of the C<sub>ant</sub> inventories (Müller et al., 2023), a signal that should be revisited to estimate the transport of C<sub>ant</sub> from the Indian Ocean towards the South Atlantic Ocean through the Agulhas Current and associated eddies or transported back to the Southern Indian Ocean through the Agulhas Return Current (Lo Monaco et al., 2005; Evans et al., 2017; Orselli et al., 2019).

#### CRediT authorship contribution statement

**Nicolas Metzl:** Writing – original draft, Methodology, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Claire Lo Monaco:** Investigation, Funding acquisition, Data curation. **Guillaume Barut:** Formal analysis. **Jean-François Ternon:** Writing – review & editing, Funding acquisition, Conceptualization.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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**Appendix A. Supplementary data**

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.dsr2.2025.105459>.

#### Data availability

All data, including new data, used are available in public data-bases (SOCAT, GLODAP, NCEI/OCADS)

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