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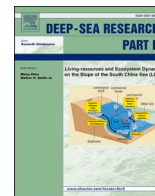
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## Distribution and long-term change of the sea surface carbonate system in the Mozambique Channel (1963–2019)

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### ABSTRACT

We report new oceanic carbonate system observations obtained during two cruises conducted in January 2004 (OISO-11) and April 2019 (CLIM-EPARSEs) in the Mozambique Channel and estimate the long-term trend of sea surface fugacity of CO<sub>2</sub> (fCO<sub>2</sub>) and pH using historical data. While in January 2004 the region was a large CO<sub>2</sub> source, the ocean was near equilibrium in April 2019. Although this region experienced a dramatic cyclone event “Idai” in March 2019 leading to low salinity and low dissolved inorganic carbon (C<sub>T</sub>) and total alkalinity (A<sub>T</sub>) concentrations in the central channel, salinity normalized A<sub>T</sub> were unchanged and C<sub>T</sub> concentrations were higher in 2019 compared to 2004 by about 12 μmol.kg<sup>-1</sup>, likely due to anthropogenic CO<sub>2</sub> uptake over 15 years. Compared to fCO<sub>2</sub> observations of 1963 in the channel, the oceanic fCO<sub>2</sub> was higher in 2004/2019 by about 100 μatm, an increase close to that observed in the atmosphere (90 ppm). A part of the fCO<sub>2</sub> increase from 1963 to 2019 (about +10 μatm) is due to the long-term ocean warming in this region (+0.011 °C.decade<sup>-1</sup>). We estimated a mean decrease of −0.087 (±0.007) pH unit between 1963 and 2019, typical of the preindustrial versus modern change in the global ocean. Using other observations in the southern part of the Mozambique Channel (around 25°S) we estimated a pH trend of −0.0129.decade<sup>-1</sup> (±0.0042) for 1963–1995 and −0.0227.decade<sup>-1</sup> (±0.0048) for 1995–2019 suggesting a strengthening of acidification trend in the Mozambique Channel in agreement with the anthropogenic CO<sub>2</sub> forcing. For the recent period, these rates were confirmed by reconstructed fCO<sub>2</sub> and pH monthly fields using a neural network model. We noted however that the pH trend in the Mozambique Channel appeared lower than previous estimates at the scale of the Indian Ocean. Based on historical atmospheric CO<sub>2</sub> data we estimated that pH in the Mozambique Channel was about 8.18 (±0.014) in the year 1800, i.e. 0.13 higher than in 2019. The concentration of C<sub>T</sub> in the year 1800 was likely around 1915 (±10) μmol.kg<sup>-1</sup>. These results will contribute to a better understanding of the impacts of ocean acidification on coral reefs since the industrial revolution by (1) providing a reference level for the reconstruction of pH from coral core samples that were collected at different locations in this region in 2019 and (2) by informing environmental authorities aiming at preserving and protecting those threatened ecosystems.

### 1. Introduction

Since the industrial revolution, about 675 GtC of anthropogenic carbon dioxide (CO<sub>2</sub>) has been emitted into the atmosphere (Friedlingstein et al., 2019) leading to an unprecedented growth of atmospheric CO<sub>2</sub> concentrations that reached on average 410 ppm in 2019 (Dlugokencky and Tans, 2020). Since 1750 about 25% of anthropogenic CO<sub>2</sub>

has been absorbed by the ocean (Friedlingstein et al., 2019) and about 31% during the period 1994–2007 (Gruber et al., 2019a). This absorption helps to mitigate global warming but induces ocean acidification (Doney et al., 2009; Feely et al., 2009; Wu et al., 2018). The latter is a major threat to marine ecosystems (Fabry et al., 2008; Gattuso et al., 2015). It impacts both calcifying organisms such as coccolithophores (Riebesell et al., 2000; Beaufort et al., 2011), foraminifera (de Moel

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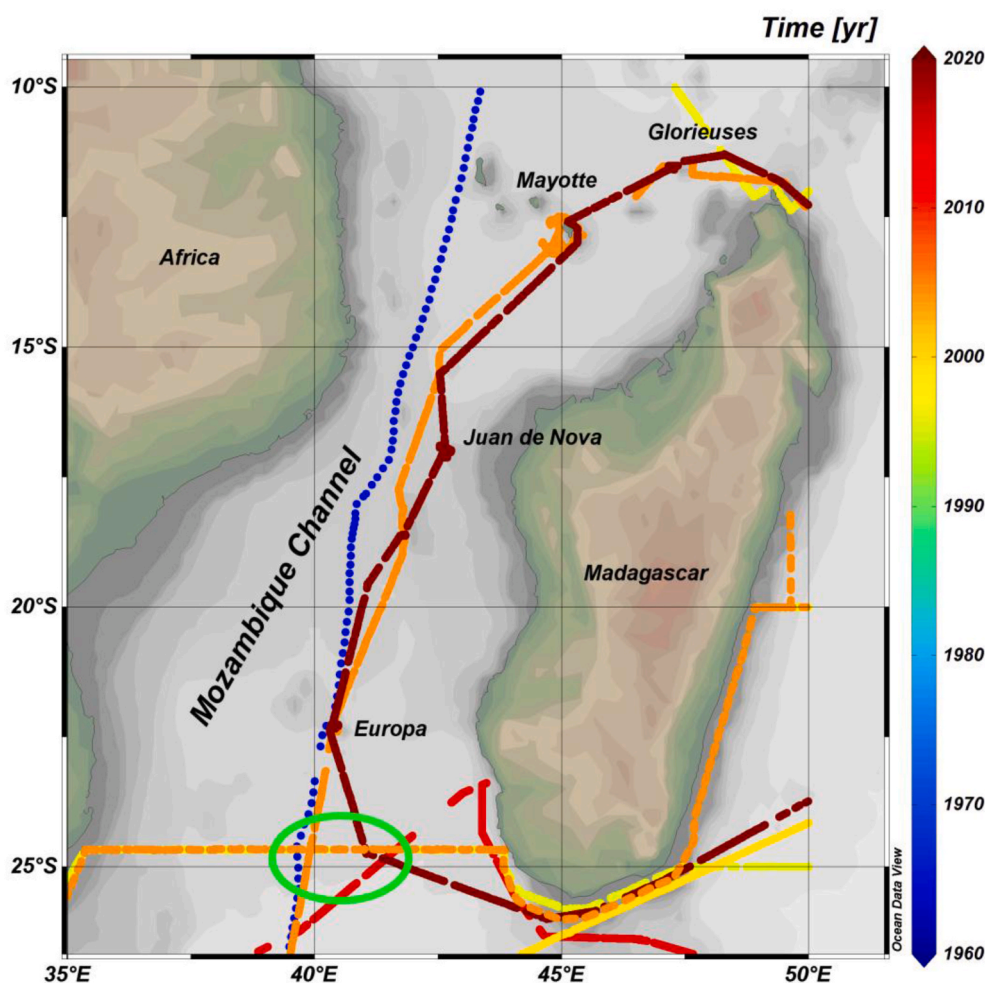
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et al., 2009), bivalves (Waldbusser et al., 2015; Tan and Zheng, 2020), and corals (Kleypas et al., 1999; Mollica et al., 2018), and major agents of carbonate dissolution, the bioeroding microflora and sponges (Schönberg et al., 2017). The accumulation of anthropogenic CO<sub>2</sub> in the ocean has led to a global decrease in pH in surface waters by on average  $-0.1$  units since the industrial revolution (Jiang et al., 2019). It is projected to further decrease by on average  $-0.2$  to  $-0.4$  units by the end of the century depending on the anthropogenic emission scenario and locations (Orr et al., 2005; Jiang et al., 2019; IPCC, 2019). Although future pH changes appear to be more pronounced in the cold and high latitudes (especially in the Arctic) compared to equatorial upwelling areas, significant pH changes are also likely to occur in the tropics and subtropics (Jiang et al., 2019; Ono et al., 2019).

Long-term observations at fixed open ocean monitoring stations since the 1980s or the 1990s show that ocean pH declined between  $-0.0013$  and  $-0.0026$  units per year depending on the location (Bates et al., 2014). High rates of surface ocean acidification are reported for coastal zones, ranging from  $-0.0020$ .yr<sup>-1</sup> ( $\pm 0.0007$ ) off the south coast of Japan (Ishii et al., 2011) to  $-0.0028$ .yr<sup>-1</sup> ( $\pm 0.0003$ ) in the Mediterranean Sea (Kapsenberg et al., 2017). In coral reef areas, high-frequency variability (e.g. Hofmann et al., 2011; Cyronak et al., 2020) confounds the assessment of the long-term pH trend by direct measurements. However, pH time series reconstructed based on coral boron isotopic ratio ( $\delta^{11}\text{B}$ ) allow to identify a prominent ocean acidification trend in the recent decades (e.g. Liu et al., 2014; Wu et al., 2018; D'Olivo et al., 2019). A compilation of direct pCO<sub>2</sub> observations at several coral reef locations (Cyronak et al., 2014) suggests that over 1992–2012 sea water pCO<sub>2</sub> increased at a rate of  $+6.6$  ( $\pm 1.4$ )  $\mu\text{atm.yr}^{-1}$ ,

i.e. up to times 3.5 faster than in the atmosphere and the open ocean. At constant temperature and alkalinity this would lead to a fast pH decline of  $-0.0055$ .yr<sup>-1</sup> in coral reef areas against  $-0.0018$ .yr<sup>-1</sup> on average in the open ocean (Feely et al., 2009; Lauvset et al., 2015; Iida et al., 2020) ultimately triggering net dissolution of reef structures (Eyre et al., 2018; Tribollet et al., 2019). At global scale, based on high quality surface ocean fCO<sub>2</sub> data compiled in the SOCAT data product version 2 (Bakker et al., 2014; Pfeil et al., 2013), and combined with regional alkalinity/salinity relationships (Lee et al., 2006), Lauvset et al. (2015) estimated rates of pH declines ranging from  $-0.0010$ .yr<sup>-1</sup> to  $-0.0027$ .yr<sup>-1</sup> in different basins over the period 1991–2011. The fastest pH decline of  $-0.0024$ .yr<sup>-1</sup> ( $\pm 0.0004$ ) for the period 1981–2011 and of  $-0.0027$ .yr<sup>-1</sup> ( $\pm 0.0005$ ) over 1991–2011 occurred in the Indian Ocean. In addition to ocean warming, such a rapid pH change, if confirmed by independent measurements and correlated to a rapid change of the carbonate saturation state, might put Indian Ocean coral reef ecosystems at risk, including many of those in the Mozambique Channel (e.g. Eparses Islands). In this oceanic region, *in situ* observations remain scarce as very few oceanographic campaigns have been conducted.

Here we present the first temporal observations of the sea surface carbonate system in the Mozambique Channel (including fugacity of CO<sub>2</sub>, fCO<sub>2</sub>, total alkalinity, A<sub>T</sub> and dissolved inorganic carbon, C<sub>T</sub>) measured during two cruises conducted in January 2004 and April 2019. We also use historical fCO<sub>2</sub> observations to explore the long-term change in fCO<sub>2</sub> and pH in this region over the period 1963–2019 and compare these variations with estimates of regional anthropogenic CO<sub>2</sub> concentrations and with reconstructed monthly pCO<sub>2</sub> and pH fields derived from a neural network model (Denvil-Sommer et al., 2019; Chau



**Fig. 1.** Cruises in the Mozambique Channel included to the SOCAT data product (version v2020, [www.socat.info](http://www.socat.info), Bakker et al., 2016, 2020). Color code is for Year. In the Mozambique Channel, cruises for May 1963 (in blue), January 2004 (OISO-11, in orange), April 2019 (CLIM-EPARSEs, in brown). Water-column data from GLODAP at 25°S were collected in Jun-1995 (yellow) and Dec-2003 (orange). All cruises used in this study are listed in Table 1. Figure produced with ODV (Schlitzer, 2013).

et al., 2020).

## 2. Data collection and methods

### 2.1. Observations during the 2004 and 2019 cruises

In January 2004 (Cruise OISO-11, 8-Jan/7-Feb 2004) and April 2019 (project CLIM-EPARSEs, 5–30 April 2019) we conducted two cruises in the Mozambique Channel on-board R.V. Marion-Dufresne (Fig. 1, Table 1). During these cruises, underway continuous surface ocean measurements were made for temperature (SST), salinity (SSS), fugacity of CO<sub>2</sub> (fCO<sub>2</sub>), total alkalinity (A<sub>T</sub>) and total dissolved inorganic carbon (C<sub>T</sub>). In addition, discrete sea surface samples were taken for the analysis of Chl-a and nutrients. In April 2019, the sampling scheme was completed by hydrocasts at selected stations (0–200m in coastal waters nearby the Eparses Islands, and 0–1000m in open waters between islands). With the exception of Chl-a, analytical methods follow the protocol used since 1998 during OISO cruises onboard R.V. Marion-Dufresne in the Southern Indian Ocean. They have been previously described (Jabaud-Jan et al., 2004; Metzl et al., 2006; Metzl, 2009).

Sea surface temperature (SST) and salinity (SSS) were measured continuously using a SBE45 thermosalinograph. Salinity data were controlled by regular sampling and conductivity measurements (Guideline Autosal 8400B and using IAPSO standard/OSIL). Sea surface temperature and salinity were also checked against CTD's surface records. Accuracies of SST and SSS are about 0.005 °C and 0.01.

In addition to the continuous underway measurements, we regularly sampled surface water for Chlorophyll-a (Chl-a) and nutrients (Nitrate and Silicate). The Chl-a samples were stored at –80 °C on-board after filtration and measured back in the laboratory using a fluorometric method (Aminot and K  rouel, 2004). Nitrate and silicate were measured onboard in January 2004 with an automatic colorimetric Technicon analyser following the methods described by Tr  guer and Le Corre (1975).

Total alkalinity (A<sub>T</sub>) and total dissolved inorganic carbon (C<sub>T</sub>) were measured on-board for both underway continuous surface and water-column samples using a potentiometric titration method (Edmond, 1970) in a closed cell. The system is automatic for sampling continuously surface seawater and transferring it into the cell. For water-column

**Table 1**

List of cruises in the Mozambique Channel (10°S–25°S) used in this study for surface fCO<sub>2</sub> data (SOCAT, version v2020, Bakker et al., 2016, 2020) and for A<sub>T</sub>/C<sub>T</sub> water column at 25°S (GLODAPv2.2019, Olsen et al., 2019).

Data-Base	Expocode	Period	Region	PI or Reference
		Month-Year	(Lat. Band)	
<b>Surface Underway fCO<sub>2</sub></b>				
SOCAT-v2020	31AR19630216	May-1963	10°S-27°S	Keeling and Waterman (1968)
SOCAT-v2020	316N19950611	Jun-1995	at 25°S	Key R
SOCAT-v2020	49NZ20031209	Dec-2003	at 25°S	Murata A.
SOCAT-v2020	35MV20040106	Jan-2004	10°S-27°S	Metzl (2009) and this study <sup>(a)</sup>
SOCAT-v2020	06BE20140710	Jul-2014	at 25°S	Steinhoff, T., Koertzing, A
SOCAT-v2020	35MV20190405	Apr-2019	10°S-27°S	This Study <sup>a</sup>
<b>Hydrocast stations (with C<sub>T</sub> and A<sub>T</sub> in water column)</b>				
GLODAPv2-2019	316N19950611	Jun-1995	at 25°S	Sabine et al. (1999)
GLODAPv2-2019	49NZ20031209	Dec-2003	at 25°S	Murata et al. (2010)

(a) In January 2004 and April 2019 A<sub>T</sub> and C<sub>T</sub> were also measured underway.

measurements, samples were collected from Niskin bottles in 500 ml glass bottles for analysis within 4h or by adding 300 µl of saturated mercuric chloride solution before storage in a cool place for subsequent analysis. For calibration, we used the Certified Referenced Materials (CRMs, Batch #52, 58 and 62 for OISO-11 and Batch #173 for CLIM-EPARSEs) provided by Pr. A. Dickson (SIO, University of California). For both cruises, we estimated the accuracy at about 3 µmol kg<sup>-1</sup> for both A<sub>T</sub> and C<sub>T</sub> (based on CRMs measurements). During CLIM-EPARSEs standard errors of 122 replicates for surface samples were ±2.6 µmol kg<sup>-1</sup> for A<sub>T</sub> and ±2.4 µmol kg<sup>-1</sup> for C<sub>T</sub>. For OISO-11, a cruise that took place in January–February 2004 in the Mozambique Channel and the Southern Indian ocean (including 15 stations), errors of replicates for deep samples (n = 19) was ±2.7 µmol kg<sup>-1</sup> for A<sub>T</sub> and ±3.3 µmol kg<sup>-1</sup> for C<sub>T</sub>. Errors of 31 replicates for surface samples were ±1.5 µmol kg<sup>-1</sup> for A<sub>T</sub> and ±1.8 µmol kg<sup>-1</sup> for C<sub>T</sub>. The water column data for OISO-11 have been quality controlled in CARINA (Lo Monaco et al., 2010) and revisited in GLODAPv2 (Key et al., 2015; Olsen et al., 2016) with no corrections applied for A<sub>T</sub> and C<sub>T</sub>. Surface underway A<sub>T</sub> and C<sub>T</sub> data for OISO-11 and CLIM-EPARSEs are both available at NCEID/OCADS (Metzl and Lo Monaco, 2018; Lo Monaco et al., 2020b).

During CLIM-EPARSEs in April 2019, one station located at 22.30°S/40.40°E was sampled near Europa Island. At 1000m, our measurements for A<sub>T</sub> and C<sub>T</sub> data are respectively 2334 µmol kg<sup>-1</sup> and 2247 µmol kg<sup>-1</sup>. We are not aware of any biogeochemical stations (including A<sub>T</sub> and C<sub>T</sub> data) in the central Mozambique Channel. However a WOCE line was sampled in the southern part at 25°S in June 1995 (Sabine et al., 1999) and reoccupied in December 2003 (Murata et al., 2010). These data are part of the GLODAPv2.2019 data product (Olsen et al., 2019). At 25°S around 1000m GLODAPv2.2019 data were in a range of 2310–2340 µmol kg<sup>-1</sup> for A<sub>T</sub> and 2200–2255 µmol kg<sup>-1</sup> for C<sub>T</sub>. Our measurements at 1000m at 22.30°S in 2019 are thus in the higher range of concentrations observed at 25°S in 1995 or 2003, not taking into account the natural variability and anthropogenic signals.

For fCO<sub>2</sub> measurements, sea-surface water was continuously equilibrated with a “thin film” type equilibrator thermostated with surface seawater (Poisson et al., 1993). The CO<sub>2</sub> in the dried gas was measured with a non-dispersive infrared analyser (NDIR, Siemens Ultramat 5F or 6F). Standard gases for calibration (around 270, 350 and 480 ppm) and atmospheric CO<sub>2</sub> were measured every 6 h. To correct xCO<sub>2</sub> dry measurements to fCO<sub>2</sub> *in situ* data, we used polynomials given by Weiss and Price (1980) for vapour pressure and by Copin-Mont  gut (1988, 1989) for temperature (temperature in the equilibrium cell was on average 0.73 °C warmer than SST during OISO-11 and 0.097 °C warmer during CLIM-EPARSEs). The oceanic fCO<sub>2</sub> for both cruises are available in SOCAT data product (version v2020, Bakker et al., 2016, 2020) and at NCEID/OCADS (Metzl, 2018; Lo Monaco et al., 2020a). Note that when added to SOCAT, original fCO<sub>2</sub> data are recomputed (Pfeil et al., 2013) using temperature correction from Takahashi et al. (1993). Given the small difference between SST and equilibrium temperature, the fCO<sub>2</sub> data from our cruises are identical (within 1 µatm) in SOCAT and NCEID/OCADS. For coherence with other cruises we therefore used fCO<sub>2</sub> values as provided by SOCAT.

Atmospheric xCO<sub>2</sub> measured on-board was on average 377.02 (±0.73) ppm in January 2004 and 410.90 (±1.40) ppm in April 2019 in the Mozambique Channel (Supp. Fig. 1). This corresponds to an increase of +33.9 ppm in 15 years, coherent with global atmospheric xCO<sub>2</sub> values. Our measurements on board in this specific region (10°S–26°S) are almost identical to the global averaged marine surface monthly mean xCO<sub>2</sub> of 377.04 ppm for January 2004 and 411.06 ppm for April 2019 (Dlugokencky and Tans, 2020). The relatively large standard-deviation of atmospheric xCO<sub>2</sub> encountered in the Mozambique Channel (up to ± 1.4 ppm in 2019) compared to other OISO cruises in the Southern Indian Ocean (Metzl, 2009) is likely linked to the terrestrial signal from Africa and/or Madagascar depending on the season and wind fields. However given the increase of atmospheric xCO<sub>2</sub> over 15 years, the observed regional atmospheric xCO<sub>2</sub> variability

during each cruise had no significant impact on the air-sea CO<sub>2</sub> disequilibrium that we explored in this study. We thus used the mean xCO<sub>2</sub> values for each cruise (377 and 411 ppm) and converted xCO<sub>2</sub> to fCO<sub>2</sub> at 100% humidity following Weiss and Price (1980) for air-sea disequilibrium estimates.

## 2.2. Other observations and data

In order to investigate the long-term variability of fCO<sub>2</sub> and pH we used all fCO<sub>2</sub> data available in the Mozambique Channel from the SOCAT data product (version v2020, Bakker et al., 2016, 2020): our observations for 2004 and 2019 and the only other cruise conducted in the Channel in May 1963 (Keeling and Waterman, 1968). These three cruises offer a first view of fCO<sub>2</sub> variations over several decades in the Mozambique Channel, between 14°S and 25°S. However, because they are representative of different seasons (January, April and May) we included fCO<sub>2</sub> data collected in the southern part of the Channel at around 25°S to confirm the fCO<sub>2</sub> and pH trends over decades deduced in the band 14°S–25°S. All the cruises used in this study are shown in Fig. 1 (listed in Table 1) from which we select only recommended fCO<sub>2</sub> data (with WOCE Flag 2 in SOCAT). In addition, in order to separate the natural and anthropogenic CO<sub>2</sub> signals, we used water-column data from June 1995 and December 2003 along 25°S (Sabine et al., 1999; Murata et al., 2010) quality controlled in GLODAPv2.2019 (Olsen et al., 2019). Our estimates of anthropogenic CO<sub>2</sub> (hereafter C<sub>ant</sub>) in this region will be compared to a recent evaluation of C<sub>ant</sub> inventory changes in the global ocean between 1994 and 2007 (Gruber et al., 2019a;b). To discuss the seasonality of the carbonate system and air-sea CO<sub>2</sub> fluxes in this region we will also compare our observations with the climatology produced by Takahashi et al. (2009, 2014). Finally, we will compare our results with reconstructed pCO<sub>2</sub> and pH monthly fields for the period 1985–2019 derived from a neural network model (named CMEMS-LSCE-FFNN, Denvil-Sommer et al., 2019; Chau et al., 2020).

## 2.3. Calculations of carbonate system properties and anthropogenic carbon

Based on the carbonate properties available for each cruise (fCO<sub>2</sub>, A<sub>T</sub> and C<sub>T</sub>) other carbonate system properties (like pH, [H<sup>+</sup>] or the carbonate saturation state Ω) are calculated using 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 KSO<sub>4</sub> constant from Dickson (1990). The total boron concentration is calculated according to Uppström (1974). We have tested different K1, K2 constants (Merbach et al., 1973; Hansson, 1973; Dickson and Millero, 1987) and compared measured fCO<sub>2</sub> with fCO<sub>2</sub> calculated from A<sub>T</sub>/C<sub>T</sub> pairs collocated in space and time (Supp. Figs. S2a and b). Given the uncertainties associated to the measurements of temperature (0.005 °C), salinity (0.01), A<sub>T</sub> and C<sub>T</sub> (2.5 μmol kg<sup>-1</sup>), the error on calculated fCO<sub>2</sub> is ±13 μatm. This error is generally larger than the mean differences observed between measured and calculated fCO<sub>2</sub> (Supp. Fig. S2b). For example, the average differences between measured and calculated fCO<sub>2</sub> when using K1, K2 constants from Lueker et al. (2000) were -5.3 (std ±4.9) μatm for 154 co-located samples in January 2004, and +4.5 (std ±5.2) μatm for 268 co-located samples in April 2019. In line with previous recommendations (Dickson et al., 2007; Orr et al., 2015) we used constants from Lueker et al. (2000) for all calculations. To account for the effect of salinity on A<sub>T</sub> and C<sub>T</sub> concentrations these properties are also normalized at constant salinity (35) following N-A<sub>T</sub> = A<sub>T</sub> \* 35/SSS and N-C<sub>T</sub> = C<sub>T</sub> \* 35/SSS. In this study we are interested in evaluating fCO<sub>2</sub> variations over several decades. As fCO<sub>2</sub> is highly dependent on SST, fCO<sub>2</sub> will be also normalized at constant temperature (29 °C) following the relation established by Takahashi et al. (1993).

$$fCO_{29} = fCO_{2SST} * EXP(0.0423*(29-SST)) \quad (1)$$

For anthropogenic CO<sub>2</sub> calculations in the water column (C<sub>ant</sub>) we used the TrOCA method (Touratier et al., 2007) that was previously applied and compared to other methods in the Southern Indian Ocean along the 32°S section (Álvarez et al., 2009). In short, given A<sub>T</sub>, C<sub>T</sub>, Oxygen (O<sub>2</sub>) and potential temperature (θ) observations at each depth level, the C<sub>ant</sub> concentrations is derived from the evolution of the quasi-conservative tracer TrOCA compared to its pre-industrial value (see Touratier et al., 2007 for a full description). The final expression used to calculate C<sub>ant</sub> is:

$$C_{ant} = \frac{O_2 + 1,279 \left( C_T - \frac{1}{2} A_T \right) - e^{\left[ \frac{7,511 - (1,087 \cdot 10^{-2}) \cdot \theta - \frac{7,81 \cdot 10^5}{A_T^2} \right]}{1,279}} \quad (2)$$

To compare with the observed C<sub>T</sub> long-term trend in surface waters and given the uncertainty on C<sub>ant</sub> of ±6.25 μmol kg<sup>-1</sup> using the TrOCA method (Touratier et al., 2007) our interpretations in the Mozambique Channel region will be limited to the upper layers (150–200m). In the Southern Indian subtropical region C<sub>ant</sub> concentrations are generally less than 10 μmol kg<sup>-1</sup> below 1000m (Sabine et al., 1999; Álvarez et al., 2009) and C<sub>ant</sub> accumulation between 1994 and 2007 less than 10 μmol kg<sup>-1</sup> below 500 m (Murata et al., 2010; Gruber et al., 2019a).

## 2.4. Neural network reconstruction of the surface ocean carbonate system

Surface ocean partial pressure of carbon dioxide (pCO<sub>2</sub>) is obtained from an ensemble-based feed-forward neural network (FFNN) model described in Denvil-Sommer et al. (2019). For this study, we used results of the latest version of the model (Chau et al., 2020) which was developed as part of the Copernicus Marine Environment Monitoring Service (CMEMS). The mean pCO<sub>2</sub> of a 100 member ensemble is used for comparison and trend analysis throughout this study. Surface ocean pH is computed from reconstructed pCO<sub>2</sub> and alkalinity with the speciation software CO2sys-MATLAB\_V2.05 (Van Heuven et al., 2011; Orr et al., 2018). The multivariate linear regression model LIAR (Carter et al., 2016; 2018) is used to derive time and space varying surface ocean alkalinity fields as a function of sea surface temperature and salinity, as well as climatological monthly mean nitrate and dissolved silica concentrations from the World Ocean Atlas 2013 version 2 (Garcia et al., 2014; www.nodc.noaa.gov/OC5/woa13/). Global surface ocean pCO<sub>2</sub> and pH reconstructions at 1° × 1° resolution start in 1985 and are updated annually in phase with yearly releases of SOCAT data. Here we used the SOCAT version v2020 (Bakker et al., 2020) and the model fields calculated for the period 1985–2019. For a full description, access to the data and a statistical evaluation of pCO<sub>2</sub> and pH reconstructions please refer to Chau et al. (2020).

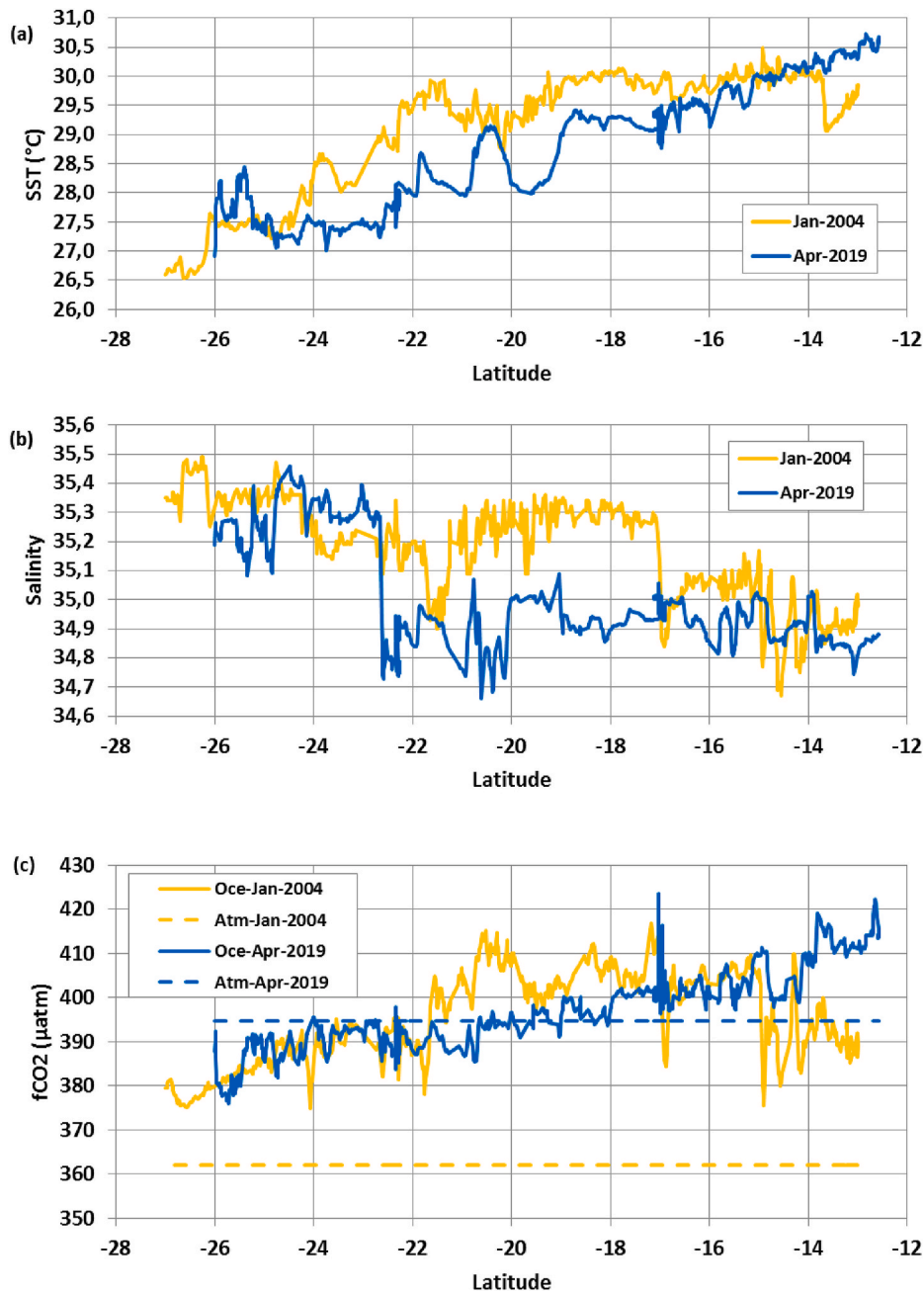
## 3. Results

### 3.1. Surface properties observed in January 2004 and April 2019 in the Mozambique Channel

#### 3.1.1. Distributions along the Mozambique Channel

Here we first present the description of sea surface observations obtained in January 2004 and April 2019 in the Mozambique Channel. Underway measurements of SST, SSS and fCO<sub>2</sub> are shown in Fig. 2 while A<sub>T</sub> and C<sub>T</sub> observations are presented in Fig. 3. Averaged values in the band 14–25°S are listed in Table 2.

During both cruises SST presents a gradual increase from south to north (Fig. 2a). SST was generally higher in January 2004 than in April 2019 except in the South (26°S) and in the North (14°S) of the Channel. In January 2004, SST reached the maximum value of 30 °C at 22°S and was quite homogeneous in the band 14°S–22°S, whereas in April 2019, SST increased more gradually to reach 30.5 °C north of Madagascar. We observed large SST and SSS variability in the central part of the Channel



**Fig. 2.** Sea surface temperature (a), salinity (b) and fCO<sub>2</sub> (c) in the Mozambique Channel observed in January 2004 (orange) and April 2019 (blue). In (c) the dashed lines correspond to atmospheric fCO<sub>2</sub>. All underway data, including near Islands are shown.

(18°S–22°S) probably linked to eddies occurring regularly in this region (Halo et al., 2014; Hancke et al., 2014; TERNON et al., 2014). This was well identified from one drifting buoy migrating southwards with an anticyclonic eddy in March–April 2019 (Supp. Fig. S3). The SST and SSS variability in the central part of the Channel is also recognized in  $A_T$  and  $C_T$  distributions (Fig. 3) but not in fCO<sub>2</sub> (Fig. 2c) because the effects of  $A_T$  and  $C_T$  on fCO<sub>2</sub> offset each other.

During each cruise, we observed a sharp front of salinity in the central part of the Mozambique Channel that was not associated to a change in SST: in January 2004 the front was located around 17°S (near Juan de Nova Island) whereas it was found at 22.5°S in April 2019 (south of Europa Island). Although the salinity was about the same in 2004 and 2019 in the South and the North of the channel, it was very different in the central region (17°S–23°S, Fig. 2b). The low salinity (SSS < 35) observed in April 2019 was linked to an excess of precipitations

that occurred in March during the dramatic tropical cyclone event called “Idai” (monthly anomaly of precipitation up to 150 mm in the band 18°S–22°S, Supp. Fig. S4). This signal was confirmed by an Argo float (Platform Code 2902142) that recorded salinity as low as 34 in the surface layer in this region in late March 2019 (Supp. Fig. S5).

Precipitations caused by the cyclone Idai directly impacted the concentrations of  $A_T$  and  $C_T$  which were thus much lower in April 2019 in the central part of the channel (Fig. 3a and b), although one would expect an increase in  $C_T$  over 15 years due to the accumulation of anthropogenic CO<sub>2</sub>. The salinity fronts at 17°S or 22.5°S (depending on the cruise) were also well identified in  $A_T$  and  $C_T$  (Fig. 3). The salinity normalized  $A_T$  ( $N-A_T$ ) and  $C_T$  ( $N-C_T$ ) distributions present much less spatial variability than  $A_T$  and  $C_T$  and are remarkably homogeneous in the Mozambique Channel south of 14°S during each cruise (Fig. 4).

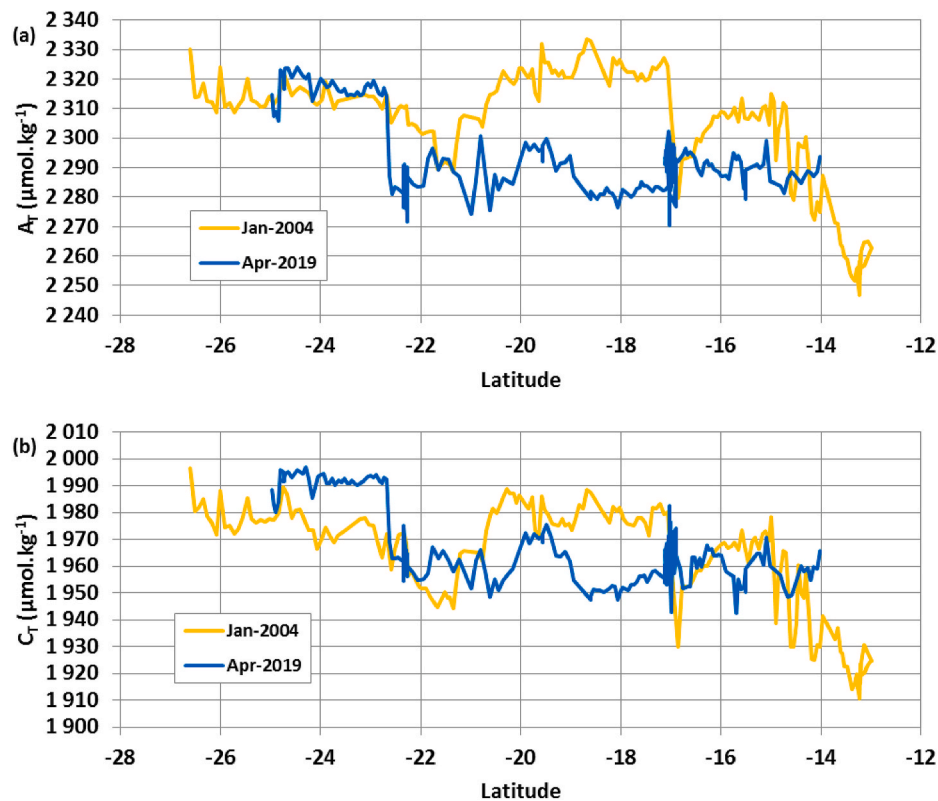


Fig. 3. (a) Sea surface  $A_T$  ( $\mu\text{mol.kg}^{-1}$ ) and (b) sea surface  $C_T$  ( $\mu\text{mol.kg}^{-1}$ ) in the Mozambique Channel observed in January 2004 (orange) and April 2019 (blue). All underway data, including near Islands are shown.

Table 2

Mean measured, calculated and trends of surface properties in the band  $14^\circ\text{S}$ – $25^\circ\text{S}$  based on observations in May 1963, January 2004 and April 2019 in the Mozambique Channel. Nb is the number of data (ND=No Data). Standard deviations are indicated in bracket. Average  $N-C_T$  and  $N-A_T$  measured only in 2004 and 2019 are also listed to compare with  $N-C_T$  and  $N-A_T$  calculated with  $f\text{CO}_2$  and  $A_T$ /SSS relationship. Adjusted values for April 2004 and April 1963 as specified were used to estimate the trends of  $f\text{CO}_2$  and  $N-C_T$  for different periods. Errors for the trends (in bracket) are based on  $f\text{CO}_2$ ,  $A_T$  and  $C_T$  measurements uncertainty for each cruise as documented in the main text and Table S1 in Supplementary Material. Last line is the  $f\text{CO}_2$  trend without the long-term warming of  $+0.011^\circ\text{C.yr}^{-1}$ .

Period	Nb	SST °C	$f\text{CO}_2$ $\mu\text{atm}$	$N-C_{T\text{cal}}$ $\mu\text{mol/kg}$	$N-A_{T\text{cal}}$ $\mu\text{mol/kg}$	Nb	$N-C_{T\text{mes}}$ $\mu\text{mol/kg}$	$N-A_{T\text{mes}}$ $\mu\text{mol/kg}$
April-2019	1677	28.61 (0.83)	397.2 (7.9)	1964.9 (4.4)	2293.1 (1.3)	376	1967.5 (6.9)	2292.8 (4.8)
January-2004	480	29.45 (0.73)	398.9 (8.9)	1958.0 (5.8)	2294.7 (1.3)	142	1955.8 (8.6)	2296.3 (5.8)
May-1963	59	25.77 (1.45)	296.2 (4.9)	1925.7 (10.0)	2292.4 (1.0)	ND	ND	ND
April-2004 (adjusted)			370.8	1949.9			1952.2	
April-1963 (adjusted)			308.1	1908.4			ND	
Trend			$f\text{CO}_2$ $\mu\text{atm.yr}^{-1}$	$N-C_{T\text{cal}}$ $\mu\text{mol.kg}^{-1}.\text{yr}^{-1}$			$N-C_{T\text{mes}}$ $\mu\text{mol.kg}^{-1}.\text{yr}^{-1}$	
2004–2019			+1.75 (0.81)	+1.00 (0.81)			+1.04 (0.79)	
1963–2004			+1.53 (0.28)	+1.00 (0.39)			ND	
1963–2019			+1.58 (0.18)	+1.00 (0.29)			ND	
1963–2019 (without warming)			+1.44 (0.18)					

### 3.1.2. Differences in $A_T$ and $C_T$ between 2004 and 2019

The largest difference in  $N-C_T$  between 2004 and 2019 is observed in the southern part of the channel ( $21^\circ\text{S}$ – $25^\circ\text{S}$ ) where  $N-C_T$  concentrations were  $10$ – $20 \mu\text{mol kg}^{-1}$  higher in April 2019 compared to January 2004 whereas  $N-A_T$  values were almost identical for both periods (Fig. 4, Table 2). Given that  $C_T$  (and  $N-C_T$ ) climatological annual cycles present higher concentrations in January than in April in the subtropical southern Indian Ocean including the Mozambique Channel (Bates et al., 2006; Takahashi et al., 2014, Supp Fig. S6), the  $N-C_T$  average increase of  $+11.8 \mu\text{mol kg}^{-1}$  observed between 2004 and 2019 (mean trend of  $0.8 \mu\text{mol kg}^{-1}.\text{yr}^{-1}$ ) likely underestimates the anthropogenic signal. Taking into account the seasonality (based on the  $C_T$  climatology computed by Takahashi et al., 2014, Supp Fig. S6),  $C_T$  would be  $16.9 \mu\text{mol kg}^{-1}$  lower

in April 2004 than in January 2004 ( $N-C_T$  in April 2004 would be  $1952.2 \mu\text{mol kg}^{-1}$  on average) and the annual increase from April 2004 to April 2019 would be  $+1.04 (\pm 0.79) \mu\text{mol.kg}^{-1}.\text{yr}^{-1}$  (Table 2). This number is in the range of the theoretical  $C_T$  trend of  $+1.2 \mu\text{mol kg}^{-1}.\text{yr}^{-1}$  calculated by assuming that surface ocean  $f\text{CO}_2$  follows the atmospheric growth rate observed over 2004–2019 ( $+2.2 \text{ppm.yr}^{-1}$ ). This suggests that most of the increase in  $N-C_T$  is due to the accumulation of anthropogenic  $\text{CO}_2$ , which could also explain part of the changes in  $f\text{CO}_2$  and pH observed between 2004 and 2019, as well as the long-term trends investigated back to the sixties in section 3.2.

### 3.1.3. $f\text{CO}_2$ variability, seasonality and biological processes

Although the  $C_T$  and  $A_T$  distributions along the Mozambique Channel

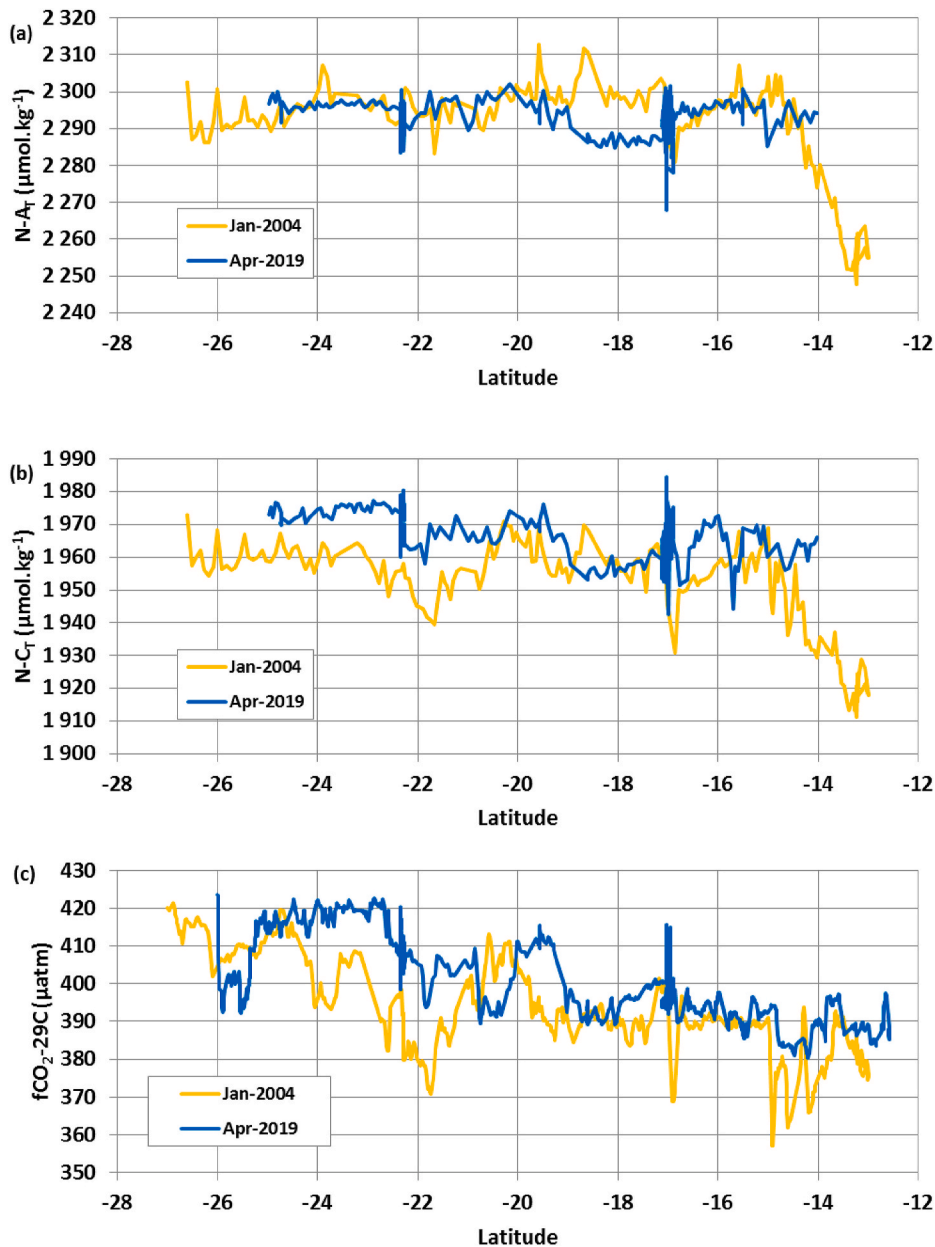


Fig. 4. Same as Fig. 3 for (a) Salinity normalized  $A_T$  ( $N-A_T$ ,  $\mu\text{mol.kg}^{-1}$ ) and (b) Salinity normalized  $C_T$  ( $N-C_T$ ,  $\mu\text{mol.kg}^{-1}$ ) in the Mozambique Channel observed in January 2004 (orange) and April 2019 (blue). Also presented in (c) the  $f\text{CO}_2$  normalized at SST  $29^\circ\text{C}$  ( $f\text{CO}_2-29\text{C}$ ,  $\mu\text{atm}$ ). All underway data, including near Islands are shown.

present large gradients associated with the salinity fronts at  $22.5^\circ\text{S}$  or  $17^\circ\text{S}$  (Fig. 3), their impact on  $f\text{CO}_2$  cancel each other and the fronts are not recognized in  $f\text{CO}_2$  measurements (Fig. 2c). In April 2019,  $f\text{CO}_2$  increased progressively northward like SST, leading to a positive  $f\text{CO}_2/\text{SST}$  relationship, except for measurements conducted in the vicinity of Juan de Nova near  $17^\circ\text{S}$  (Supp. Fig. S7). In January 2004, the  $f\text{CO}_2$  distribution presented a larger spatial variation leading to a poor relation between  $f\text{CO}_2$  and SST (Supp. Fig. S7), notably due to the low  $f\text{CO}_2$  values (below  $390\ \mu\text{atm}$ ) and high SST observed north of  $15^\circ\text{S}$  and around  $22^\circ\text{S}$  where we also measured minima in  $C_T$  (Fig. 3b) and in  $N-C_T$  (Fig. 4b). These signals were linked to locally enhanced biological activity revealed in both surface Chl-a *in-situ* measurements around  $15^\circ\text{S}$  and  $22^\circ\text{S}$  in January 2004 (Supp. Fig. S8) and at regional scale in the monthly Chl-a distribution derived from MODIS (Supp. Fig. S8). In April 2019, *in-situ* Chl-a concentrations were in the same range as in January 2004 with few local maxima also detected around  $15^\circ\text{S}$  and  $22^\circ\text{S}$  (Supp.

Fig. S8) and also associated with minima in  $C_T$  and  $N-C_T$  (Figs. 3b and 4b). In the vicinity of the islands (Europa at  $22^\circ\text{S}$  and Juan de Nova at  $17^\circ\text{S}$ ) the Chl-a concentrations in April 2019 present some variability but in the range of observations in the open ocean ( $0\text{--}0.25\ \text{mg m}^{-3}$ ). High *in-situ* Chl-a concentrations are thus only identified at small-scale during both cruises and these local events do not impact the overall trend of the  $f\text{CO}_2$  and  $C_T$  changes in the Channel between 2004 and 2019. Instead, satellite derived Chl-a products suggest a rather homogeneous Chl-a distribution and slightly higher values in April 2019 compared to January 2004, notably in the central region (Supp. Fig. S8a). This is reflected in the mean seasonal cycle of Chl-a in this region, suggesting a slight gradual increase of Chl-a from December to July (Supp. Fig. S9). Over the region  $15^\circ\text{S}\text{--}26^\circ\text{S}$ , the average Chl-a concentration derived from MODIS was  $0.11\ \text{mg m}^{-3}$  in January 2004 and  $0.17\ \text{mg m}^{-3}$  in April 2019, a mean difference of only  $0.06\ \text{mg m}^{-3}$  not captured in our *in-situ* localized data (Supp. Fig. S8). In April 2019, the



MODIS Chl-a concentrations of  $0.17 \text{ mg m}^{-3}$  were the same as the mean climatological value for April ( $0.17 \text{ mg m}^{-3} \pm 0.02$  on average for years 2003–2019, Supp. Fig. S9). Thus, although one would expect the cyclone “Idai” that occurred in March 2019 to have impacted primary production (e.g. through input of nutrients either from the subsurface or from terrigenous material through atmospheric transport), we conclude that, except for salinity, observations in April 2019 were close to climatological conditions.

As noted above, climatological  $C_T$  concentrations in the channel are on average lower in April than in January (Takahashi et al., 2014) and the  $f\text{CO}_2$  decrease from January to April cannot be explained by temperature alone. The seasonal  $C_T$  change between winter and summer (up to  $30 \mu\text{mol kg}^{-1}$ ) is likely driven by biological activity but not directly recognized by Chl-a observations (Supp. Fig. S9). This seasonal draw-down of  $C_T$  is potentially associated to biological  $\text{N}_2$ -fixation similarly to observations in the south-western tropical Pacific where a similar  $C_T$  seasonality has been reported (Moutin et al., 2018). In the Western Indian Ocean and the Mozambique Channel *Trichodesmium* blooms ( $\text{N}_2$ -fixers) have been detected from satellite data (Westberry and Siegel, 2006). However, the occurrence of blooms from remote sensing data only remains qualitative and obviously limited to large surface aggregations of cells (McKinna, 2015). The hypothesis that diazotrophy may play a significant role in  $C_T$  and  $f\text{CO}_2$  seasonality is further supported by the observation of high abundances of *Trichodesmium* spp in the Mozambique Channel in April 2011 (Dupuy et al., 2016) but we have no *in-situ* data to document this process during the 2004 and 2019 cruises.

### 3.1.4. Difference in $f\text{CO}_2$ between 2004 and 2019

Although atmospheric  $f\text{CO}_2$  was much higher in 2019 than in 2004, the oceanic  $f\text{CO}_2$  range was similar (between 380 and 420  $\mu\text{atm}$  for both periods, Fig. 2c) as well as the mean  $f\text{CO}_2$  in the band  $14^\circ\text{S}$ – $25^\circ\text{S}$  (Table 2). In January 2004, the ocean  $f\text{CO}_2$  was much higher than in the atmosphere (a  $\text{CO}_2$  source) with  $\Delta f\text{CO}_2$  ( $\Delta f\text{CO}_2 = f\text{CO}_{2\text{oc}} - f\text{CO}_{2\text{atm}}$ ) ranging from 20 to 50  $\mu\text{atm}$ , with a mean value of  $+36.8 (\pm 8.9) \mu\text{atm}$ . In April 2019, the mean  $\Delta f\text{CO}_2$  value was much lower ( $+2.5 \pm 7.9$ )  $\mu\text{atm}$ : the ocean was a  $\text{CO}_2$  source north of  $18^\circ\text{S}$  (maximum  $\Delta f\text{CO}_2$  of  $+20 \mu\text{atm}$ ), near-equilibrium with the atmosphere in the central channel ( $22^\circ\text{S}$ – $18^\circ\text{S}$ ) and a  $\text{CO}_2$  sink south of  $25^\circ\text{S}$  (minimum  $\Delta f\text{CO}_2$  of  $-20 \mu\text{atm}$ ). Not taking into account the increase in oceanic  $f\text{CO}_2$  from 2004 to 2019 due to anthropogenic  $\text{CO}_2$  uptake, our mean results showing a  $\text{CO}_2$  source in January and near-equilibrium in April, are coherent with monthly climatological  $\Delta f\text{CO}_2$  values in this region, i.e.  $+19.95 (\pm 3.3) \mu\text{atm}$  for January and  $+1.2 (\pm 5.3) \mu\text{atm}$  for April (Takahashi et al., 2009). Since very few observations in the Mozambique Channel were used to construct the  $p\text{CO}_2$  climatology (Takahashi et al., 2009) our results validate the  $p\text{CO}_2$  extrapolation in this region that has been later used to create  $C_T$  and pH climatologies (Takahashi et al., 2014).

In the ocean the seasonal variation of  $f\text{CO}_2$  is controlled by a complex interplay of thermodynamic, biological, chemical and mixing processes (Takahashi et al., 2002). In the subtropics, including the South Indian Ocean sector, the effect of temperature is generally found to be the dominant process at seasonal (Louanchi et al., 1996; Metzl et al., 1995; Takahashi et al., 2002) and inter-annual scales (Metzl et al., 1998). In order to take into account the thermal effect, observed  $f\text{CO}_2$  in January 2004 and April 2019 were normalized to a constant temperature of  $29^\circ\text{C}$  ( $f\text{CO}_2$ -29C) close to the average SST observed in January and April. The  $f\text{CO}_2$ -29C distributions are shown in Fig. 4c. For both periods,  $f\text{CO}_2$ -29C variations tend to follow the  $\text{N}-C_T$  distributions (Fig. 4b). As opposed to  $f\text{CO}_2$ , but like for  $\text{N}-C_T$ ,  $f\text{CO}_2$ -29C was on average slightly higher in April 2019 ( $403.4 \pm 8.5 \mu\text{atm}$ ) than in January 2004 ( $391.9 \pm 10.9 \mu\text{atm}$ ). The difference of only  $+11.5 \mu\text{atm}$  is much lower than the atmospheric increase of  $+33.9 \text{ ppm}$  between 2004 and 2019. The temperature drives only part of the  $f\text{CO}_2$  monthly variations and it is thus important to correct for other seasonal contributions to evaluate long-term trends in  $f\text{CO}_2$ . If we adjust the  $f\text{CO}_2$  observed in January 2004 to April 2004 based on the climatology (Takahashi et al., 2014, Supp Fig. S6b),  $f\text{CO}_2$

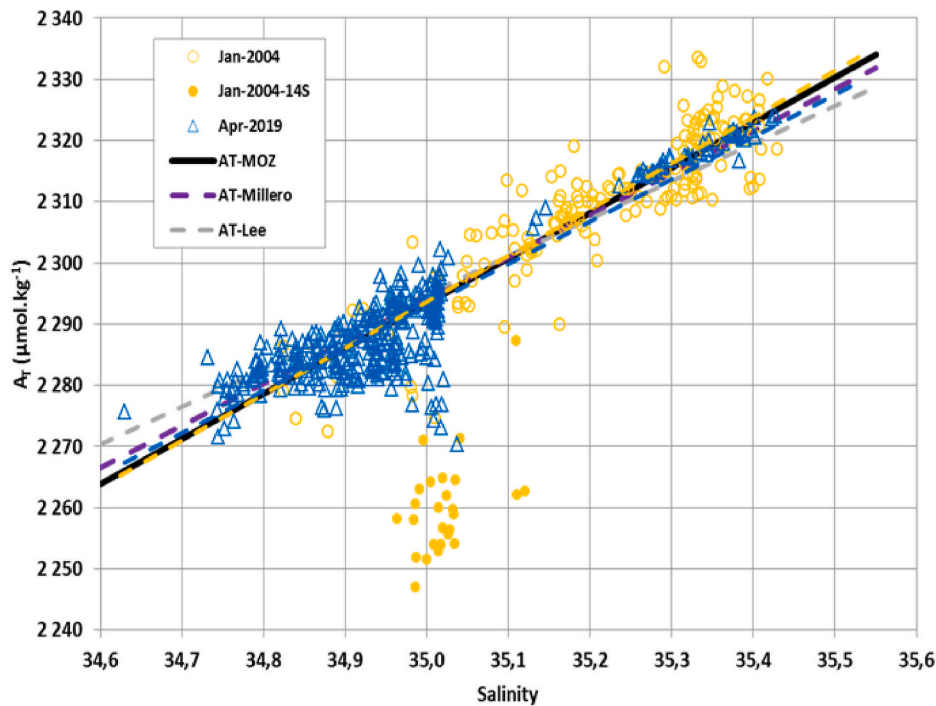
data for January are reduced by  $28.1 \mu\text{atm}$  and we obtain a mean value of  $370.8 \mu\text{atm}$  for April 2004 (Table 2). The  $f\text{CO}_2$  increase from April 2004 to April 2019 would lead to a rate of  $1.75 (\pm 0.81) \mu\text{atm.yr}^{-1}$  over the last 15 years, slower than  $+2.2 \text{ ppm.yr}^{-1}$  in the atmosphere over the same period. At constant  $A_T$  the observed increase in  $f\text{CO}_2$  would translate into a trend of  $+1.01 (\pm 0.45) \mu\text{mol.kg}^{-1}.\text{yr}^{-1}$  in  $C_T$ , the same rate deduced from  $C_T$  measurements described above ( $+1.04 \pm 0.79 \mu\text{mol kg}^{-1}.\text{yr}^{-1}$  for  $C_T$  also adjusted to April, Table 2). This simple calculation based on 2 cruises conducted 15 years apart indicates that on average  $\text{CO}_2$  concentrations increased in the Mozambique Channel at a rate of around  $1.0 \mu\text{mol kg}^{-1}.\text{yr}^{-1}$  and that seasonal signals of  $f\text{CO}_2$  and  $C_T$  have to be taken into account to properly evaluate the  $f\text{CO}_2$  and  $C_T$  trends. The same is true when one uses  $f\text{CO}_2$  data and  $A_T$ /SSS relationship to calculate pH and to evaluate the long-term signal of ocean acidification (e.g. Lauvset et al., 2015).

### 3.1.5. Selecting the regional $A_T$ /SSS relationship

Similar to what is commonly observed in the global ocean (Millero et al., 1998) there is a strong relationship between  $A_T$  and salinity for both cruises (Fig. 5), except in the vicinity of Mayotte Island in January 2004 (north of  $14^\circ\text{S}$ ). The  $A_T$ /SSS relationships are almost the same for January 2004 and April 2019 and close to  $A_T$ /SSS relationships derived at large scale in the subtropics or in the Indian Ocean (Millero et al., 1998; Lee et al., 2006). This suggests that these empirical  $A_T$ /SSS relationships could be used to reconstruct spatial and temporal distribution of  $A_T$  from salinity. In turn, pH can be calculated from reconstructed  $A_T$  and  $f\text{CO}_2$  data (when only  $f\text{CO}_2$  data are available as it is the case for SOCAT, Bakker et al., 2016). In order to select the best  $A_T$ /SSS relationship for the Mozambique Channel, we first compared several relations with  $A_T$  measurements obtained in January 2004 and April 2019 (Supp. Fig. S10). All relationships, including that of Millero et al. (1998) or Lee et al. (2006), lead to the same residuals with no statistical differences. Therefore, in the following we used the  $A_T$ /SSS relationship derived from our observations in 2004 and 2019 (black line in Fig. 5):  $A_T (\mu\text{mol.kg}^{-1}) = 73.841 (\pm 1.15) * \text{SSS} - 291.02 (\pm 40.4)$  ( $n = 548$ ,  $r^2 = 0.88$ ).

### 3.1.6. Detecting recent pH changes in the Mozambique channel

The  $A_T$ /SSS relationship described above enables to calculate pH from  $f\text{CO}_2$  and SSS underway measurements. Here we are first interested in detecting the change in pH between 2004 and 2019. As we had  $A_T$ ,  $C_T$  and  $f\text{CO}_2$  observations for both cruises, we compared pH calculated with  $A_T$  and  $C_T$  pairs or with  $f\text{CO}_2$  and  $A_T$  based on the  $A_T$ /SSS relationship. Average calculated pH values in the band  $14^\circ\text{S}$ – $25^\circ\text{S}$  are listed in Table 3. Both results are presented in Fig. 6 for pH at  $29^\circ\text{C}$  (pH-29C). Although calculated pH is subject to uncertainties due to the error in each property (SST, SSS,  $A_T$ ,  $C_T$  or  $f\text{CO}_2$ ) the overall pH-29C distributions are the same either based on  $A_T$ - $C_T$  pairs or on underway  $f\text{CO}_2$  data. Except in the vicinity of the islands pH-29C values ranged between 8.02 and 8.06. Although pH values at *in-situ* temperature were almost the same (Table 3), pH-29C values were clearly lower in April 2019 compared to January 2004 (Fig. 6, Table 3). On average pH-29C in April 2019 was lower by  $-0.0116$  or  $-0.0218$  depending on the calculation. The pH difference between the 2 cruises reflects both the seasonality and probably the pH decrease due to the accumulation of anthropogenic  $\text{CO}_2$  over 15 years. To take into account the seasonality, we adjusted the mean  $f\text{CO}_2$  and  $C_T$  observation in January 2004 to April 2004 based on the climatology (Takahashi et al., 2014) and recalculate pH adjusted to April 2004 (Table 3). The pH difference between 2019 and 2004 was  $-0.0238 (\pm 0.0018)$  (using average  $f\text{CO}_2$  data and associated errors) or  $-0.0244 (\pm 0.0050)$  (using average  $A_T$ - $C_T$  data). Over 15 years, both results lead to a trend of between  $-0.0154.\text{decade}^{-1}$  and  $-0.016.\text{decade}^{-1}$  (Table 3). Given all uncertainties attached to pH calculations, our estimates either based on  $f\text{CO}_2$  (and reconstructed  $A_T$ ) or  $A_T$ - $C_T$  observations show that acidification occurred in the Mozambique Channel at a rate similar to that reported for the northern subtropics ( $-0.017$ .



**Fig. 5.** Total Alkalinity ( $A_T$ ,  $\mu\text{mol.kg}^{-1}$ ) versus salinity in surface water in the Mozambique Channel (14–25°S) observed in January 2004 (orange circles) and April 2019 (blue triangle). Linear relation for each period are also shown (orange and blue dashed-lines). In January 2004 data north of 14°S near Mayotte are identified with filled orange circles and not used for the  $A_T$ /SSS relationship. Also shown are the  $A_T$ /SSS relationships from Millero et al. (1998) for the Indian Ocean (Purple dashed) and from Lee et al. (2006) for the subtropical oceans (Grey dashed). The  $A_T$ /SSS relationship based on Jan-2004 and Apr-2019 data in the Mozambique Channel (noted AT-MOZ, black line) is the final relationship used in this analysis:  $A_T = 73.841 * S - 291.02$ .

**Table 3**

Mean sea surface and trends of pH in the band 14°S–25°S in May 1963, January 2004 and April 2019 in the Mozambique Channel. Nb is the number of data (ND= No Data). Standard deviations for mean values and errors for trends are indicated in bracket. Errors for trends are based on  $f\text{CO}_2$ ,  $A_T$  and  $C_T$  measurements uncertainty for each cruise as documented in the main text and Table S1 in Supplementary Material. pH is calculated with  $f\text{CO}_2$  data and  $A_T$ /SSS relationship or with measured  $C_T$  and  $A_T$  (for 2004 and 2019). Averaged pH at SST-29 °C is also listed. Adjusted values for April 2004 and April 1963 as specified were used to estimate the trends of pH for different periods. Last line is the pH trend without the long-term warming of  $+0.011 \text{ } ^\circ\text{C.yr}^{-1}$ .

Period	Nb ( $f\text{CO}_2$ )	pH TS	pH-29C TS	Nb ( $A_T$ - $C_T$ )	pH TS	pH-29C TS
April-2019	1677	8.040 (0.008)	8.034 (0.007)	376	8.035 (0.008)	8.029 (0.008)
January-2004	480	8.039 (0.008)	8.045 (0.009)	142	8.044 (0.010)	8.051 (0.010)
May-1963	59	8.143 (0.008)	8.094 (0.015)	ND	ND	ND
April-2004 (adjusted)			8.063		8.059	
Avril-1963 (adjusted)			8.126		ND	
Trend		pH ( $f\text{CO}_2$ ) TS.yr <sup>-1</sup>			pH ( $A_T$ - $C_T$ ) TS.yr <sup>-1</sup>	
2004–2019		-0.00154 (0.00100)			-0.00160 (0.00149)	
1963–2004		-0.00154 (0.00039)				
1963–2019		-0.00154 (0.00028)				
1963–2019 (without warming)		-0.00137 (0.00028)				

decade<sup>-1</sup>, e.g. Bates et al., 2014; Ono et al., 2019). This is, however, much lower than previous estimates of pH trends derived from  $f\text{CO}_2$  data at large scale in the Indian Ocean. Lauvset et al. (2015) report rates between  $-0.024.\text{decade}^{-1}$  for the period 1981–2011 and  $-0.027.$

decade<sup>-1</sup> for 1991–2011 which suggests a faster pH decrease in the Indian Ocean compared to other regions. The pH trend of  $-0.00160.\text{yr}^{-1}$  ( $\pm 0.00149$ ) derived in our observations in the Mozambique Channel between 2004 and 2019 is also close to the average trend of  $-0.00165.\text{yr}^{-1}$  ( $\pm 0.00038$ ) computed over 2004–2019 in the same region from reconstructed monthly pH fields using a neural-network method (Denvil-Sommer et al., 2019; Chau et al., 2020).

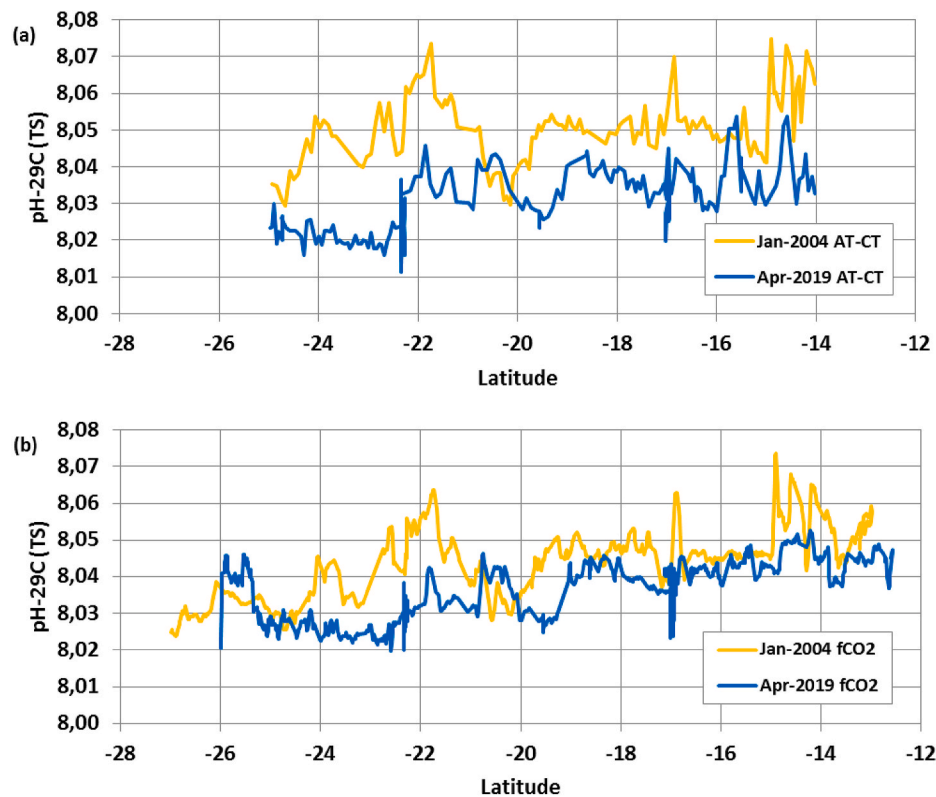
The carbonate system parameters ( $A_T$ ,  $C_T$ ,  $f\text{CO}_2$ ) measured in January 2004 and April 2019 described in this section, along with pH estimates using different inputs ( $A_T$  and  $C_T$  or  $f\text{CO}_2$ ), show that (i) N- $A_T$  concentrations are stable, (ii) there is a well characterized  $A_T$ /SSS relationship, (iii) the changes in  $C_T$ ,  $f\text{CO}_2$  and pH over 15 years are significant and (iv) when seasonality is taken into account the observed trends of  $C_T$ ,  $f\text{CO}_2$  and pH likely reflect the uptake of anthropogenic  $\text{CO}_2$ . In the following section, we explore these signals over a longer time period extending back to the sixties.

### 3.2. Detecting the long-term trends of $f\text{CO}_2$ and pH in the Mozambique channel

#### 3.2.1. Comparison with historical observations collected in the Mozambique channel in 1963

In order to explore the long-term change in  $f\text{CO}_2$  and pH in the Mozambique Channel, we compared our  $f\text{CO}_2$  observations obtained in 2004 and 2019 with the only other  $f\text{CO}_2$  dataset obtained in this region along a latitudinal transect in May 1963 (Keeling and Waterman, 1968). To estimate pH changes, we used  $f\text{CO}_2$ , SST and SSS data for the 3 cruises and the  $A_T$ /SSS relation described above. We note that in May 1963, salinity was not measured and for this cruise we used the mean monthly salinity from the World Ocean Atlas, WOA (Antonov et al., 2006), as listed in the SOCAT data product (Bakker et al., 2016). The mean WOA salinity in the band 14°S–25°S in May is 34.88 close to those measured in January 2004 (35.15) and April 2019 (34.97). The use of WOA salinity has a negligible impact on pH calculations and derived trends. The comparison of observations obtained in 1963, 2004 and 2019 is presented in Fig. 7 for the original data and in Fig. 8 for  $f\text{CO}_2$  and pH normalized at SST 29 °C along with calculated N- $C_T$  for these cruises.

The atmospheric  $x\text{CO}_2$  value in May 1963 was around 315 ppm, 90



**Fig. 6.** Surface water pH (at SST 29 °C) distribution in the Mozambique Channel observed in January 2004 (orange) and April 2019 (blue). (a) pH at 29 °C calculated with  $A_T$  and  $C_T$  pairs. (b) pH at 29 °C calculated with  $fCO_2$  and  $A_T$  reconstructed from salinity. Data very close to Islands filtered (e.g. Juan de Nova at 17°S).

ppm lower than in 2019. Like in April 2019, oceanic  $fCO_2$  in May 1963 was quite homogeneous in the Mozambique Channel increasing progressively northward with values ranging from 290  $\mu atm$  at 25°S to 305  $\mu atm$  at 16°S (Fig. 7b). This region was a small  $CO_2$  sink during this period. Average  $\Delta fCO_2$  was  $-9.3 (\pm 4.9) \mu atm$  in May 1963, very close to the climatological value for May ( $-5.3 \pm 7.4 \mu atm$  for a reference year 2000, Takahashi et al., 2009) and not very different from our recent observations in April 2019 (near equilibrium with average  $\Delta fCO_2$  of  $+2.5 \pm 7.9 \mu atm$ ). The most remarkable change is, like for atmospheric  $CO_2$ , the much lower ocean  $fCO_2$  observed in May 1963 (Fig. 7b, Table 2). Consequently this leads to higher pH in 1963 compared to 2004 or 2019 (Fig. 7c, Table 3). Based on these data, oceanic  $fCO_2$  increased by  $+101 (\pm 5) \mu atm$  from 1963 to 2019 and pH decreased by  $-0.104 (\pm 0.006)$ . This change in pH is similar to the reduction estimated for the global ocean between preindustrial and modern times of  $-0.11$  (Jiang et al., 2019). These differences are a snapshot between two cruises conducted 56 years apart and modulated by SST variations that should be taken into account to better interpret the long-term trends of  $fCO_2$  and pH.

### 3.2.2. Seasonal temperature and the impact of long-term warming on the $fCO_2$ trend

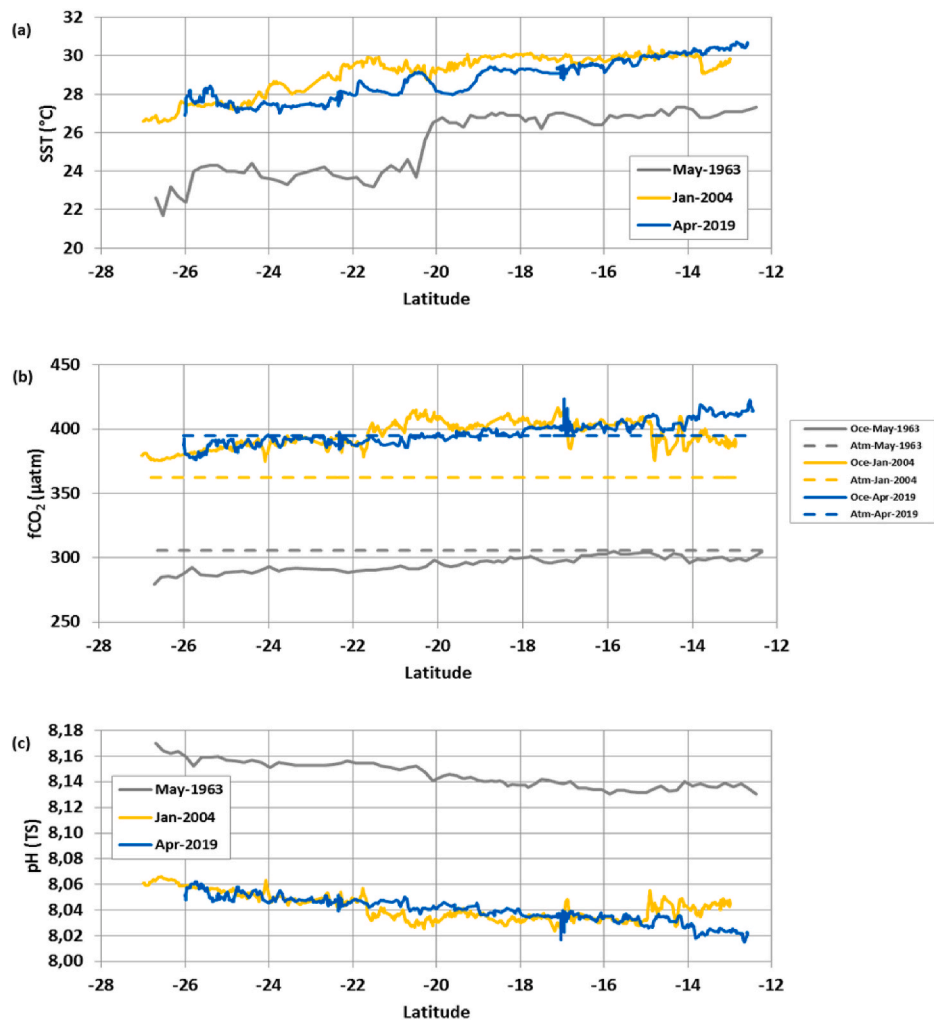
The observed SST seasonal variation is significant with the ocean being much colder in May 1963 compared to January 2004 or April 2019 (Fig. 7a). The measurements in 1963 present also a sharp front in SST at 20°S which is not seen in  $fCO_2$  records (Fig. 7b). This is probably because both  $A_T$  and  $C_T$  concentrations in May 1963 (not measured in 1963) drive  $fCO_2$  the same way as observed in April 2019 (see description above, Fig. 2). In 1963, the increase in SST from 24 °C in the south to 27 °C in the north would lead to a  $fCO_2$  increase of about  $+45 \mu atm$  but measurements show only a difference of  $+15 \mu atm$  from south to north. This suggests a competitive balance between the thermal, physical and biological processes, all leading to rather homogeneous  $fCO_2$  and pH distributions in 1963 (Fig. 7b and c).

In addition, the Indian Ocean, including the Mozambique Channel, experienced a significant long-term warming in recent decades that would drive part of the  $fCO_2$  and pH trends. Based on monthly SST products (Reynolds et al., 2002) extracted at 15°S–20°S in the Mozambique Channel from 1981 to 2019 we estimated an annual long-term warming of  $+0.011 (\pm 0.007) ^\circ C.yr^{-1}$  (Supp. Fig. S11). Such a pronounced warming has also been identified in the Indian Ocean subtropics ( $>0.1 ^\circ C.decade^{-1}$ ) dating back to the sixties (Alory et al., 2007; Roxy et al., 2014). It could be due to a fast response to recent climate change or to natural multi-decadal variability (Zinke et al., 2014). Relying on corals sampled in the reefs off Southwestern Madagascar at 43°E–23°S, Zinke et al. (2014) reconstructed a warming rate of  $+0.13 ^\circ C.decade^{-1}$  between the years 1720 and 1800 and between 1900 to present.

A long-term warming around  $+0.1 ^\circ C.decade^{-1}$  is also consistent with the observed SST in May 1963 in the band 14°S–20°S ( $26.82 \pm 0.25 ^\circ C$ ) being  $0.7 ^\circ C$  lower than the mean climatological SST in May computed in recent years ( $27.52 \pm 0.39 ^\circ C$  for the period 2000–2018). This long-term warming would increase sea surface  $fCO_2$  by about  $+7.5 \mu atm$  and would decrease pH by  $-0.009$  units, a signal not directly linked to the oceanic  $CO_2$  uptake. This corresponds to about 7% of the net  $fCO_2$  increase of  $+101 \mu atm$  observed between 1963 and 2019.

### 3.2.3. Changes in $fCO_2$ , pH and $C_T$ in the Mozambique channel over 56 years

To separate the effect of temperature and anthropogenic  $CO_2$ ,  $fCO_2$  and pH were normalized at SST 29 °C (Fig. 8a and b). We also compared  $N-C_T$  calculated with  $fCO_2$  and reconstructed  $A_T$  for the 3 cruises (Fig. 8c, Table 2). To validate the pH and  $N-C_T$  calculations based on  $fCO_2$  data for 1963, we first compared the  $N-C_T$  calculated with  $fCO_2$  ( $N-C_{Tcal}$ ) and reconstructed  $A_T$  with  $N-C_T$  derived from the measurements ( $N-C_{Tmes}$ ) described in Section 3.1 for 2004 and 2019. On average and taking into account the uncertainty of the measurements and calculations, the  $N-C_{Tcal}$  and  $N-C_{Tmes}$  values were of the same order for each cruise



**Fig. 7.** Surface water temperature (a),  $f\text{CO}_2$  (b) and calculated pH (c) in the Mozambique Channel observed in May 1963 (grey), January 2004 (orange) and April 2019 (blue). In (b) atmospheric  $f\text{CO}_2$  also shown for each year (same color, dashed line).

(Table 2). In 2019 both  $N\text{-C}_{\text{Tcal}}$  and  $N\text{-C}_{\text{Tmes}}$  were higher than in 2004 (respectively +7 and +12  $\mu\text{mol kg}^{-1}$ ) and adjusted values for April lead to the same  $N\text{-C}_T$  trend for 2004–2019 (Table 2). We have thus confidence in  $N\text{-C}_T$  and pH values calculated with  $f\text{CO}_2$  and the  $A_T/\text{SSS}$  for 1963.

For all three cruises,  $f\text{CO}_2\text{-}29\text{C}$  and  $N\text{-C}_T$  decreased northward and the opposite for  $\text{pH}\text{-}29\text{C}$  (Fig. 8). In May 1963 the decrease occurred more sharply at the SST front near  $20^\circ\text{S}$  (Fig. 7a). Consequently, the changes in  $f\text{CO}_2$ , pH and  $N\text{-C}_T$  between 1963 and 2004/2019 are slightly more pronounced in the North. The same contrast is observed for aragonite ( $\Omega_{\text{Ar}}$ ) and calcite ( $\Omega_{\text{Ca}}$ ) saturation states calculated with  $f\text{CO}_2$  and  $A_T/\text{SSS}$  for each cruise (Supp. Fig. S12).

On average  $f\text{CO}_2$  and  $N\text{-C}_T$  in 1963 were much lower than in 2004 and 2019, and  $\text{pH}\text{-}29\text{C}$  much higher (Fig. 8, Tables 2 and 3). To estimate the long-term trend,  $f\text{CO}_2$  observations in May 1963 were adjusted to April 1963 (+11.9  $\mu\text{atm}$  based on the climatology) and compared with April 2004 and 2019 to calculate the trends (Tables 2 and 3). From 1963 to 2004, the  $f\text{CO}_2$ ,  $N\text{-C}_T$  and pH trends were respectively +1.5 ( $\pm 0.3$ )  $\mu\text{atm}\cdot\text{yr}^{-1}$ , +1.0 ( $\pm 0.4$ )  $\mu\text{mol}\cdot\text{kg}^{-1}\cdot\text{yr}^{-1}$  and  $-0.0015\cdot\text{yr}^{-1}$  ( $\pm 0.0004$ ). From 1963 to 2019 results are almost the same, +1.6 ( $\pm 0.2$ )  $\mu\text{atm}\cdot\text{yr}^{-1}$ , +1.0 ( $\pm 0.3$ )  $\mu\text{mol}\cdot\text{kg}^{-1}\cdot\text{yr}^{-1}$  and  $-0.0015\cdot\text{yr}^{-1}$  ( $\pm 0.0003$ ).

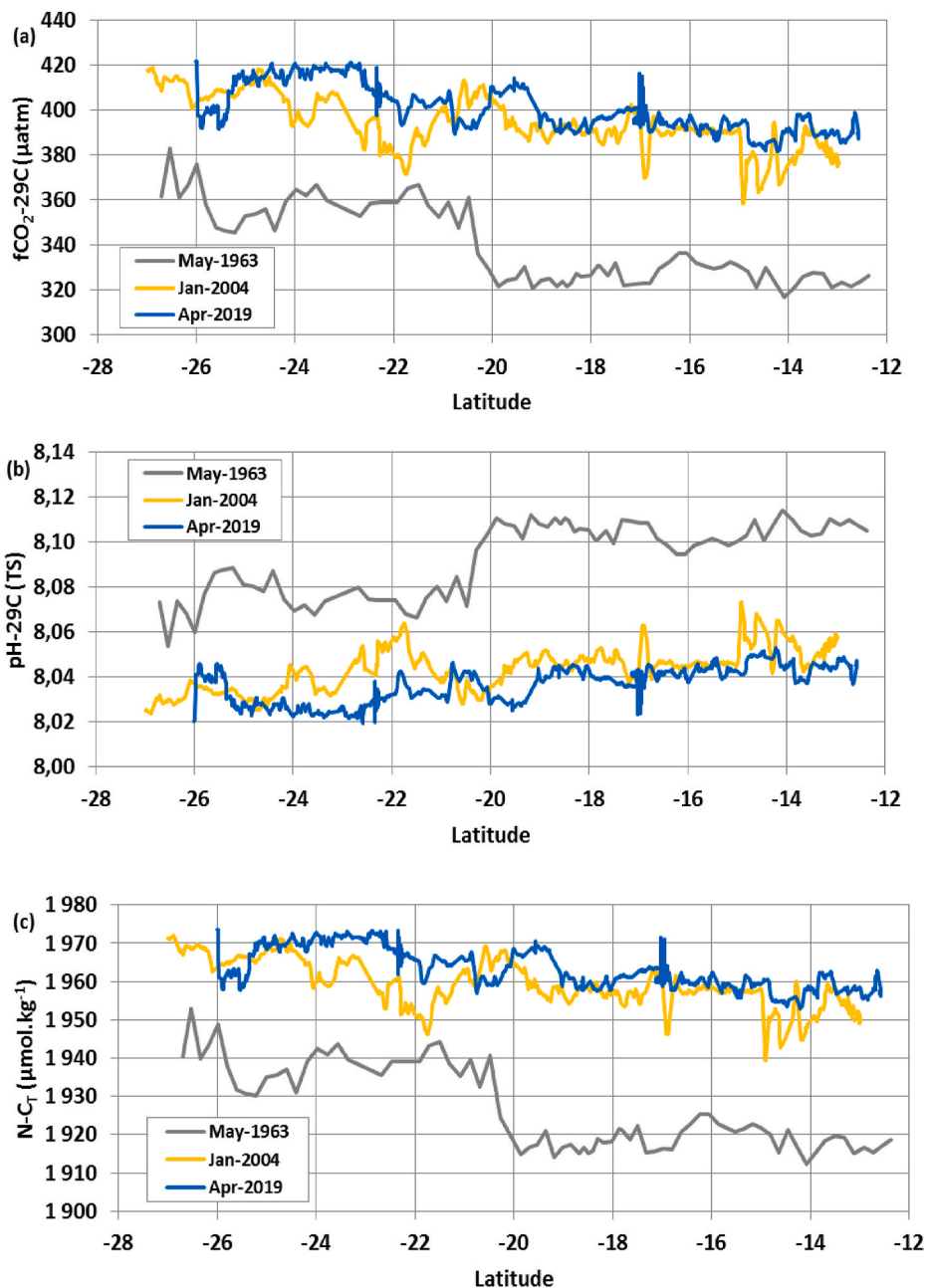
These results, although based on only three cruises, are remarkably close to the atmospheric  $x\text{CO}_2$  trend of +1.6  $\text{ppm}\cdot\text{yr}^{-1}$  over 1963–2019 and to theoretical trends of +1.04  $\mu\text{mol kg}^{-1}\cdot\text{yr}^{-1}$  for  $\text{C}_T$  and  $-0.00165\cdot\text{yr}^{-1}$  for pH assuming that the ocean followed the atmospheric  $\text{CO}_2$

increase and constant alkalinity. Subtracting the effect of the long-term warming in this region (+0.011  $^\circ\text{C}\cdot\text{yr}^{-1}$ ) on the  $f\text{CO}_2$  trend (+0.13  $\mu\text{atm}\cdot\text{yr}^{-1}$ ), leads to a trend of  $f\text{CO}_2$  of 1.39  $\mu\text{atm}\cdot\text{yr}^{-1}$  (1963–2004) or 1.44  $\mu\text{atm}\cdot\text{yr}^{-1}$  (1963–2019) that would reflect the anthropogenic  $\text{CO}_2$  uptake. Our results also suggest that observed trends in the Mozambique Channel were slightly more pronounced in the recent decades (Tables 2 and 3), but this is deduced from only three cruises. For a better evaluation of the decadal variability and long-term trends we now focus on the southern region of the Mozambique Channel where more  $f\text{CO}_2$  data are available for different seasons and years (Table 1).

### 3.3. Long-term trends in the southern region of the Mozambique channel

To confirm the changes observed in the Mozambique Channel from the three cruises in 1963, 2004 and 2019, we explored the  $f\text{CO}_2$  and pH variations based on all data available in SOCAT-v2020 around  $25^\circ\text{S}$ . Here we selected the data in the region  $23.5^\circ\text{S}\text{-}26.5^\circ\text{S}/38^\circ\text{E}\text{-}42^\circ\text{E}$  (identified with a green circle in Fig. 1). This added 3 cruises conducted in June 1995, December 2003 and July 2014 (Table 1). In 1995 and 2003  $\text{C}_T$  and  $A_T$  were also measured at fixed stations (Sabine et al., 1999; Murata et al., 2010) and we used these surface data (3–10m) to compare with  $N\text{-C}_T$  calculated from  $f\text{CO}_2$  and  $A_T$ .

Sea surface temperature,  $f\text{CO}_2$  and pH (calculated) for the six cruises are shown in Fig. 9 and the averages of observed and calculated properties around  $25^\circ\text{S}$  for each cruise are listed in Table 4. For each period



**Fig. 8.** Surface water  $f\text{CO}_2$  at  $\text{SST} = 29^\circ\text{C}$  (a) and calculated pH at  $\text{SST} = 29^\circ\text{C}$  (b) in the Mozambique Channel observed in May 1963 (grey), January 2004 (orange) and April 2019 (blue). In (c) also shown  $\text{N-C}_T$  distribution calculated with  $f\text{CO}_2$  and  $A_T$  reconstructed.

$\text{SST}$ ,  $f\text{CO}_2$  and pH are fairly homogeneous in this region (Fig. 9) and this translates into a low standard-deviations around the mean (Table 4). The ocean was colder in May, June and July than in December, January and April (Fig. 9a). For the cold season as expected, the  $f\text{CO}_2$  was the lowest in May 1963 ( $<300 \mu\text{atm}$ ) compared to June 1995 ( $>300 \mu\text{atm}$ ) and July 2014 ( $>350 \mu\text{atm}$ ) and pH was higher in 1963 ( $>8.15$ ) compared to 2014 ( $<8.09$ ). During the warm season (December–April),  $f\text{CO}_2$  was always higher than  $370 \mu\text{atm}$  (Fig. 9b). As described above, the same  $f\text{CO}_2$  values (near  $390 \mu\text{atm}$ ) were observed in January 2004 and April 2019, i.e. 15 years apart. The lowest pH (range  $8.033 < \text{pH} < 8.066$ ) was always observed during the warm season (Fig. 9c).

The two cruises conducted one month apart in December 2003 and January 2004 on different ships recorded surprisingly the same range of  $f\text{CO}_2$ , around  $385\text{--}390 \mu\text{atm}$  (Table 4). At  $40^\circ\text{E}$ , however, despite  $\text{SST}$  being the same (Fig. 9a),  $f\text{CO}_2$  was higher in December 2003 than in January 2004 by  $+12 \mu\text{atm}$  on average (Fig. 9b). This is opposed to the

climatology according to which December  $f\text{CO}_2$  is generally lower by  $-20 \mu\text{atm}$  than January in this region (Supp. Fig. S6). An inspection of the Quality Control information available on-line in SOCAT ([www.socat.info](http://www.socat.info)) indicated that for the December 2003 cruise (Expocode 49NZ20031209) there was no equilibrator temperature and pressure recorded. We are thus less confident with the accuracy of  $f\text{CO}_2$  data for this cruise (Lauvset et al., 2019). Like for our cruises in 2004 and 2019, the  $\text{C}_T$  concentrations measured in 1995 and 2003 were close to the  $\text{C}_T$  calculated from  $f\text{CO}_2$  and our  $A_T/\text{SSS}$  relationship (last two columns in Table 4) confirming the pH values calculated the same way for different years and seasons. For the trend analysis we thus use pH and  $\text{C}_T$  calculated using  $f\text{CO}_2$  data for all cruises (except December 2003).

The temporal evolutions of  $f\text{CO}_2$ , pH and  $\text{N-C}_T$  averaged at  $25^\circ\text{S}$  are shown in Fig. 10 and the trends evaluated on different periods listed in Table 5 and illustrated in Fig. 11. The trends were estimated after adjusting  $f\text{CO}_2$  data to June each year (grey diamonds in Fig. 10). We

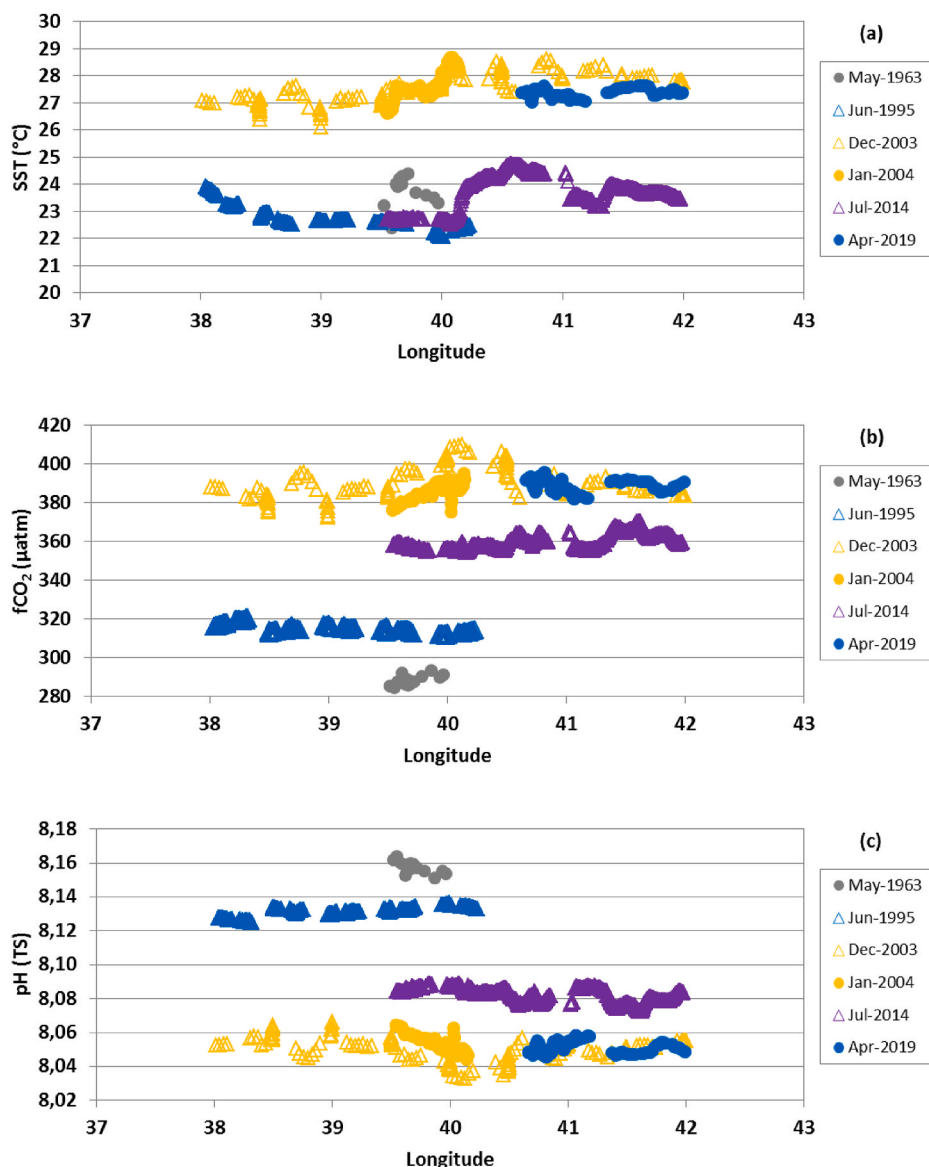


Fig. 9. Sea surface water temperature (a),  $fCO_2$  (b) and calculated pH (c) around 25°S in the Mozambique Channel observed in May 1963, June 1995, December 2003, January 2004, July 2014 and April 2019. Cruises are listed in Table 1 and tracks identified in Fig. 1. The mean observations for each cruise are listed in Table 4.

Table 4

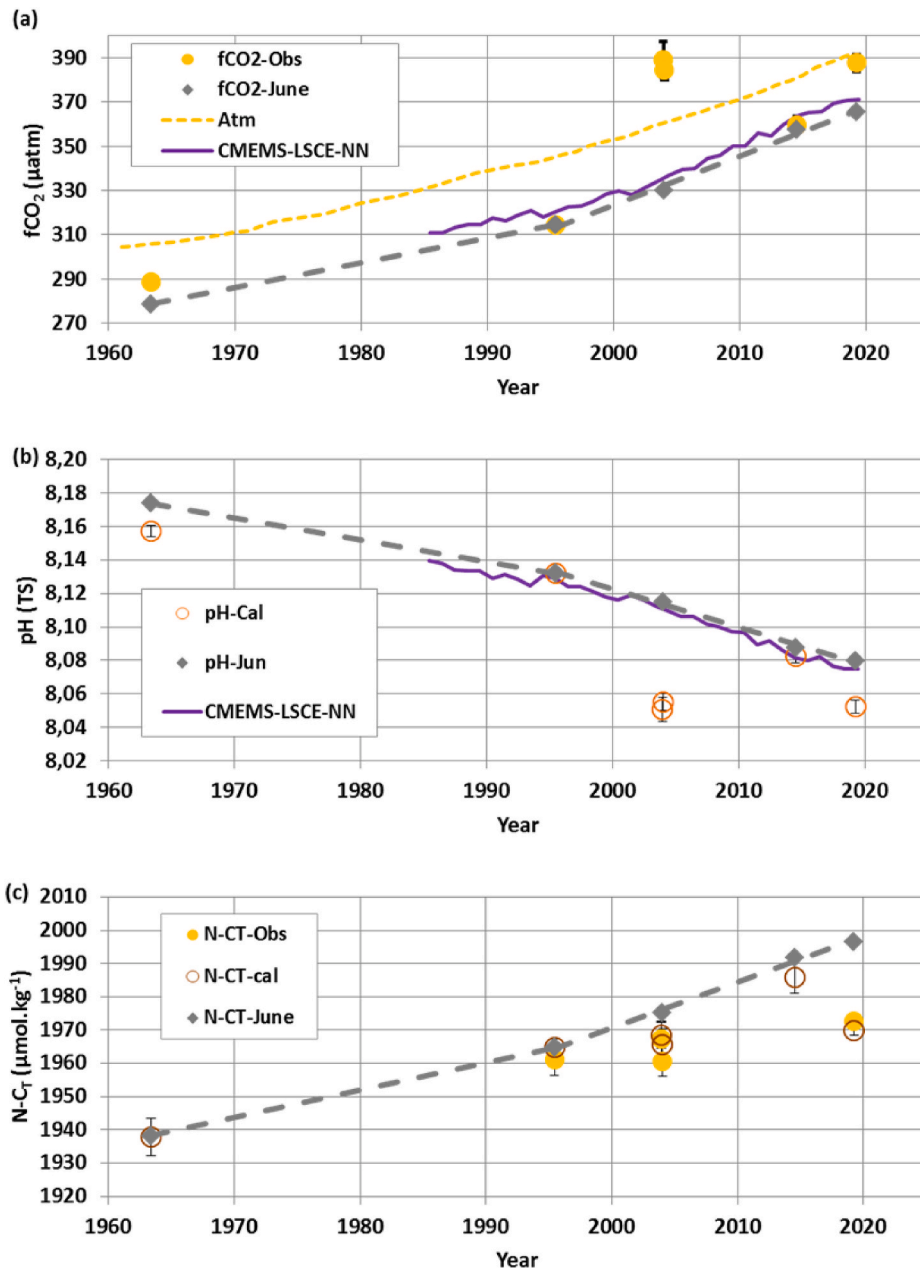
Mean sea surface measured or calculated properties around 25°S for 6 cruises in the south part of Mozambique Channel. Nb = number of data from surface underway sampling used to calculate  $[H^+]$ , pH and  $N-C_T$ . Standard deviations are in bracket.  $N-C_T$  stands for salinity normalized  $C_T$  concentrations either calculated with  $fCO_2$  and  $A_T$  ( $N-C_{Tcal}$ ) or measured ( $N-C_{Tmes}$ ) when available. ND= No Data.

Period	Nb	SST °C	$fCO_2$ µatm	$[H^+]$ nmol.kg <sup>-1</sup>	pH TS	$N-C_{Tcal}$ µmol.kg <sup>-1</sup>	$N-C_{Tmes}$ µmol.kg <sup>-1</sup>
May-1963	15	23.70 (0.59)	288.6 (2.6)	6.965 (0.055)	8.157 (0.003)	1937.8 (5.6)	ND
June-1995	717	22.66 (0.39)	314.6 (1.9)	7.381 (0.044)	8.132 (0.003)	1964.7 (2.6)	1961.3 (4.8) n9
Dec-2003	180	27.58 (0.53)	389.3 (8.1)	8.901 (0.146)	8.051 (0.007)	1968.4 (3.9)	1967.4 (5.3) n5
Jan-2004	111	27.61 (0.53)	384.6 (4.5)	8.813 (0.101)	8.055 (0.005)	1965.6 (3.3)	1960.5 (4.5) n24
Jul-2014	820	23.57 (0.67)	359.7 (3.9)	8.268 (0.080)	8.083 (0.004)	1985.9 (5.0)	ND
Apr-2019	156	27.31 (0.16)	387.8 (3.8)	8.864 (0.080)	8.052 (0.004)	1969.9 (1.4)	1972.6 (2.0) n17

also compared the observational results with the  $pCO_2$  and pH values reconstructed by Chau et al. (2020). Here we extracted the CMEMS-LSCE-NN model gridded results at the location 25°S-40°E. The trends from the model are evaluated for June to compare with observations (Fig. 12) or using the annual values (Table 5). Given the errors in the trends estimated for each property based on both measurements

uncertainty, spatial variability and pH or  $N-C_T$  calculations, trends for short periods, e.g. 1995–2004 (Fig. 11) have large errors and thus the interpretation of observed changes is mainly limited to multi-decadal variations (1963–1995 versus 1995–2019).

We first noted that the long-term trends estimated with all data at 25°S are almost identical to those derived when using only 1963 and



**Fig. 10.** Temporal evolution of  $f\text{CO}_2$  (a), pH (b) and  $\text{N-C}_T$  (c) around  $25^\circ\text{S}$  in the Mozambique Channel based on data shown in Fig. 9. Measured  $f\text{CO}_2$  and  $\text{N-C}_T$  are indicated by orange dots. pH and  $\text{N-C}_T$  calculated with  $f\text{CO}_2$  and  $A_T$  are indicated by open circles. In all panels, values adjusted to the month of June indicated by grey diamond and used for trend estimates (dashed grey lines). In (a) and (b) results from monthly reconstructed  $p\text{CO}_2$  and pH (at  $25^\circ\text{S}$ – $40^\circ\text{E}$ ) shown in purple line for June 1985–2019 (CMEMS-LSCE-NN, Chau et al., 2020). In (a) also shown the atmospheric  $f\text{CO}_2$  (orange dotted line). The mean observations for each cruise are listed in Table 4 and trends listed in Table 5. Standard-deviations for observations and calculated pH or  $\text{N-C}_T$  are indicated by vertical bars (generally about the size of symbols).

2019 observations in the Mozambique Channel for the band  $14^\circ\text{S}$ – $25^\circ\text{S}$  as described above. This suggests that the results at  $25^\circ\text{S}$  reflect the trends in a larger domain. However, these additional data allowed us to identify a shift in the trends before and after 1995, as expected from the recent faster increase of atmospheric  $\text{CO}_2$  concentrations. At  $25^\circ\text{S}$ , the trends evaluated over the period 1995–2019 were almost twice as large as those evaluated over 1963–1995 (Table 5, Fig. 11). For  $f\text{CO}_2$  we estimated an annual rate that changed from  $+1.14 (\pm 0.23) \mu\text{atm.yr}^{-1}$  in 1963–1995 to  $+2.20 (\pm 0.26) \mu\text{atm.yr}^{-1}$  in 1995–2019 (Figs. 10a and 11). For pH the trends varied from  $-0.00129 (\pm 0.00042)$  to  $-0.00227 (\pm 0.00048)$  unit  $\text{yr}^{-1}$  for the same periods (Figs. 10b and 11). To confirm these results we also evaluated the temporal change of pH based on  $[\text{H}^+]$  concentrations calculated for each cruise (Supp. Fig. S13) and using Equation (3) (Kwiatkowski and Orr, 2018).

$$\Delta\text{pH} = -1 \Delta[\text{H}^+]/2.303 [\text{H}^+] \quad (3)$$

where  $\Delta\text{pH}$  and  $\Delta[\text{H}^+]$  are the temporal differences between each period (1995 versus 1963 or 2019 versus 1995) and  $[\text{H}^+]$  the concentration at

the beginning of each period (1963 or 1995). The derived pH trends using Equation (3) are  $-0.00137 \text{ yr}^{-1}$  for 1963–1995 and  $-0.00232 \text{ yr}^{-1}$  for 1995–2019 (Table 5). Like for the trends deduced from the mean pH values at  $25^\circ\text{S}$  (Fig. 10b), the pH trend based on  $[\text{H}^+]$  concentrations is about twice faster in 1995–2019. For the recent period the observed  $f\text{CO}_2$  and pH trends are remarkably close to those derived from the CMEMS-LSCE-NN model (Fig. 10a and b, Fig. 12, Table 5). For  $\text{C}_T$ , we estimated a trend of  $+0.82 (\pm 0.41) \mu\text{mol.kg}^{-1}.\text{yr}^{-1}$  for 1963–1995 and  $+1.37 (\pm 0.42) \mu\text{mol.kg}^{-1}.\text{yr}^{-1}$  for 1995–2019 in the same range as the trends deduced when comparing data collected in 2004 and 2019 in the whole Channel (section 3.1, Table 2) and indicative of the accumulation of anthropogenic  $\text{CO}_2$ .

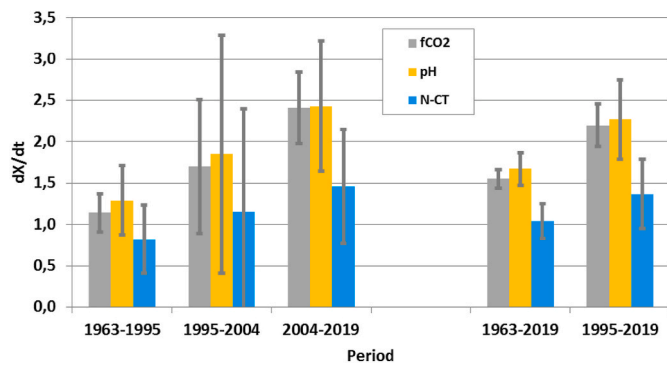
#### 3.4. Anthropogenic $\text{CO}_2$ in the southern region of the Mozambique channel

In order to separate the contributions of natural or climate change induced variations (e.g. warming) and the accumulation of

**Table 5**

Trends observed around 25°S in the Mozambique Channel evaluated from observations between 1963 and 2019 adjusted to June (as shown in Fig. 10 and Supp. Fig. S13 for  $[H^+]$ ). Standard errors of slope are in bracket. Trends for pH are also calculated from  $[H^+]$  concentrations following Equation (3) (see text, noted  $pH_{H^+}$ ). Also indicated are the trends for atmospheric  $xCO_2$  for different periods and from the monthly reconstructed  $pCO_2$  and pH values based on a neural network model (CMEMS-LSCE-NN, Denvil-Sommer et al., 2019; Chau et al., 2020). For CMEMS-LSCE-NN the trends are for June or using all months in 1985–2019. For comparison the trends in June for period 1995–2004, 2004–2019 and 1995–2019 are underlined. ND = No Data. Detail for errors on the trends is documented in the Methods and Table S2 in the Supplementary Material.

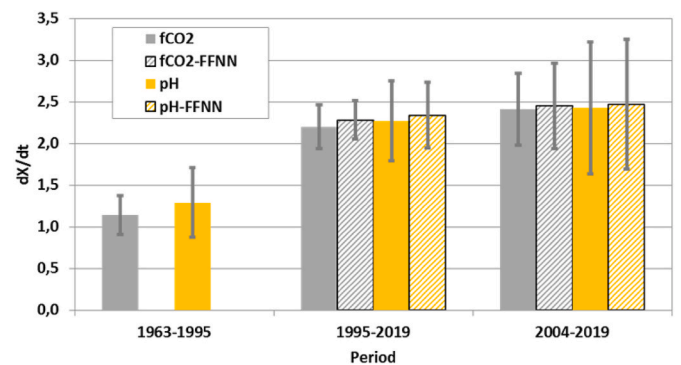
Period	Atm $xCO_2$ ppm.yr <sup>-1</sup>	$fCO_2$ $\mu atm.yr^{-1}$	$[H^+]$ nmol.kg <sup>-1</sup> .yr <sup>-1</sup>	$pH_{H^+}$ TS.yr <sup>-1</sup>	pH TS.yr <sup>-1</sup>	$N-C_{Treal}$ $\mu mol.kg^{-1}.yr^{-1}$
1963–2019	+1.60 (0.02)	+1.55 (0.11)	0.0290 (0.0036)	−0.00188	−0.00167 (0.0002)	+1.04 (0.21)
1963–1995	+1.35 (0.02)	+1.14 (0.23)	0.0213 (0.0069)	−0.00137	−0.00129 (0.00042)	+0.82 (0.41)
<u>1995–2004</u>	+1.82 (0.04)	<u>+1.70</u> (0.81)	0.0318 (0.0242)	−0.00190	<u>−0.00185</u> (0.00144)	+1.15 (1.25)
<u>2004–2019</u>	+2.13 (0.04)	<u>+2.41</u> (0.43)	0.0452 (0.0145)	−0.00247	<u>−0.00243</u> (0.00079)	+1.46 (0.69)
<u>1995–2019</u>	+2.04 (0.03)	<u>+2.20</u> (0.26)	0.0412 (0.0085)	−0.00232	<u>−0.00227</u> (0.00048)	+1.37 (0.42)
CMEMS-LSCE-NN		$fCO_2$ $\mu atm.yr^{-1}$			pH TS.yr <sup>-1</sup>	
<u>1995–2004 June</u>		<u>+1.70</u> (0.95)			<u>−0.00186</u> (0.00163)	
<u>2004–2019 June</u>		<u>+2.45</u> (0.51)			<u>−0.00247</u> (0.00078)	
<u>1995–2019 June</u>		<u>+2.28</u> (0.23)			<u>−0.00234</u> (0.00039)	
1985–2019 June		+1.89 (0.14)			−0.00196 (0.00024)	
1995–2004 Annual		+2.02 (0.48)			−0.00203 (0.00071)	
1995–2019 Annual		+1.88 (0.14)			−0.00180 (0.00019)	
2004–2019 Annual		+1.80 (0.30)			−0.00165 (0.00038)	
1985–2019 Annual		+1.80 (0.09)			−0.00177 (0.00012)	



**Fig. 11.** Annual trends (see Table 5) estimated for different periods in the southern Mozambique Channel (25°S) based on observations for  $fCO_2$  (grey,  $\mu atm.yr^{-1}$ ), pH (orange, unit.yr<sup>-1</sup>) and  $N-C_T$  (blue,  $\mu mol.kg^{-1}.yr^{-1}$ ). For pH, the annual trend is multiplied by  $-1000$  (a pH trend of 1.5 in the figure is  $-0.0015 yr^{-1}$ ). For short periods such as 1995–2004 the errors are large and thus the interpretation is limited. For longer periods,  $fCO_2$  and pH trends are higher in 2004–2019 compared to 1963–1995 and mainly linked to anthropogenic  $CO_2$  as suggested by  $N-C_T$  changes.

anthropogenic  $CO_2$  to the  $fCO_2$  and pH trends described above, we evaluated the anthropogenic fraction of  $C_T$  (hereafter noted  $C_{ant}$ ) in the southern Mozambique Channel region based on water-column observations. Data from two cruises at 25°S in June 1995 and December 2003 are available in the GLODAPv2.2019 data product (Olsen et al., 2019). Murata et al. (2010) used these observations to evaluate the changes of anthropogenic  $CO_2$  concentrations in the interior of the South Indian Ocean. They estimated a mean increase of  $C_{ant}$  of  $+7.9 (\pm 1.1) \mu mol.kg^{-1}$  from June 1995 to December 2003 in the upper layer of the subtropical water in the Central Indian Ocean. Over 8.5 years this corresponds to a trend of  $+0.93 (\pm 0.13) \mu mol.kg^{-1}.yr^{-1}$ , in the range of our estimate based on surface observations (Fig. 10c, Table 5). Specifically for the period 1995–2004, we evaluated an increase in  $N-C_T$  of  $+10.5 \mu mol.kg^{-1}$  in surface waters but over only 9 years the trend is uncertain,  $+1.15 (\pm 1.25) \mu mol.kg^{-1}.yr^{-1}$  (Table 5).

To further explore  $C_T$  and  $C_{ant}$  variations across the Mozambique Channel, we revisited the data used by Murata et al. (2010) focusing on the stations within the same region as selected for surface data described in the previous section (25°S/38°E–42°E). To calculate  $C_{ant}$  we used the



**Fig. 12.** Annual trends (see Table 5) for  $fCO_2$  (grey,  $\mu atm.yr^{-1}$ ) and pH (orange, unit.yr<sup>-1</sup>) estimated for different periods in the southern Mozambique Channel (25°S) based on observations (filled bars) and the CMEMS-LSCE-FFNN model (hatched bars). For pH, the trend is multiplied by  $-1000$  (a pH trend of 1.5 in the figure is  $-0.0015 yr^{-1}$ ). Given the errors associated to these estimates, the trends are the same either based on observations or the CMEMS-LSCE-FFNN model.

TrOCA method developed by Touratier et al. (2007) (see Section 2.3). Because indirect methods like TrOCA are not suitable to evaluate  $C_{ant}$  concentrations in surface waters (due to biological activity and gas exchange) we calculated  $C_{ant}$  in the layer 150–200m below the nitracline. The averages of observed and calculated properties in this layer are listed in Table 6 as well as the differences between the cruises. From 1995 to 2003  $C_T$  and  $N-C_T$  increased by about  $10 \mu mol.kg^{-1}$  and no significant change was observed for  $A_T$ . Our  $C_{ant}$  estimate in this region increased from  $35.1 (\pm 1.2) \mu mol.kg^{-1}$  in 1995 to  $42.3 (\pm 3.2) \mu mol.kg^{-1}$  in 2003. The  $C_{ant}$  increase of  $+7.3 (\pm 3.4) \mu mol.kg^{-1}$  reported here around 25°S is very close to the average value of  $+7.9 (\pm 1.1) \mu mol.kg^{-1}$  derived by Murata et al. (2010) in the subtropical Indian Ocean subsurface waters. The  $C_{ant}$  increase explains 70% of the  $C_T$  increase of  $+10.5 \mu mol.kg^{-1}$  over this period (1995–2003). The remaining 30% is probably linked to natural  $C_T$  variations associated to internal processes such as eddy activity around 24°S (Swart et al., 2010) and/or remineralization as revealed by the decrease in oxygen concentration (Table 6).

Our estimate of  $C_{ant}$  variations at regional scale can be also compared to the results by Gruber et al. (2019a) who evaluated the changes in  $C_{ant}$  in the global ocean between 1994 and 2007. We extracted the  $C_{ant}$  data



**Table 6**

Mean observed or calculated properties and their temporal differences in the layer 150–200m in the region 25°S/38–42°E. Data are from GLODAPv2.2019 (Olsen et al., 2019). All units in  $\mu\text{mol.kg}^{-1}$ . Standard-deviations are in bracket. Nb is number of samples in the 150–200m layer.

Month-Year	Expocode	$C_T$	$N-C_T$	$A_T$	$N-A_T$	$O_2$	$C_{\text{ant}}$	Nb
Jun-1995	316N19950611	2070.8 (5.9)	2039.0 (5.2)	2329.8 (2.6)	2293.8 (1.9)	194.8 (11.8)	35.1 (1.2)	9
Dec-2003	49NZ20031209	2081.3 (14.2)	2048.0 (15.6)	2332.8 (5.6)	2295.5 (3.1)	184.4 (14.7)	42.3 (3.2)	8
Difference 2003–1995		+10.5 (15.4)	+9.0 (16.4)	+3 (6.7)	+1.6 (3.7)	−10.4 (18.9)	+7.3 (3.4)	

for the Mozambique Channel made available by Gruber et al. (2019b) and estimated the mean  $C_{\text{ant}}$  changes at 25°S in different layers (Table 7). The accumulation of  $C_{\text{ant}}$  between 1994 and 2007 is rather homogeneous in the Mozambique Channel (Supp. Fig. S14). In the top layer (0–200m) the mean  $C_{\text{ant}}$  increase between 1994 and 2007 is +14.03 ( $\pm 0.78$ )  $\mu\text{mol.kg}^{-1}$  in the Channel (15°S–27°S). At 25°S, the accumulation in the layer 150–200m is +12.94 ( $\pm 0.56$ )  $\mu\text{mol.kg}^{-1}$ , i.e. a rate of +1.00 ( $\pm 0.04$ )  $\mu\text{mol.kg}^{-1}.\text{yr}^{-1}$  which is slightly higher than our  $C_{\text{ant}}$  estimate of +0.86 ( $\pm 0.4$ )  $\mu\text{mol.kg}^{-1}.\text{yr}^{-1}$  based on TrOCA and for the period June 1995 to December 2003.

Although the  $C_{\text{ant}}$  estimates described above were obtained for different periods and using different methods (Murata et al., 2010; Gruber et al., 2019a; this study), all results lead to the same conclusion: there was a gradual increase in  $C_T$  concentrations in the upper waters of the southern Mozambique Channel from +0.82 ( $\pm 0.41$ )  $\mu\text{mol.kg}^{-1}.\text{yr}^{-1}$  in 1963–1995 to +1.37 ( $\pm 0.42$ )  $\mu\text{mol.kg}^{-1}.\text{yr}^{-1}$  in 1995–2019 (Table 5, Fig. 11). It is mainly driven by the accumulation of anthropogenic  $\text{CO}_2$  (Tables 6 and 7). This also explains the observed long-term trends of  $f\text{CO}_2$  and the progressive pH decline in this region (Fig. 10). The warming of +0.11  $^{\circ}\text{C}.\text{decade}^{-1}$  would translate into a pH decrease of  $-0.0017.\text{decade}^{-1}$ , i.e. about 10% of the observed long-term trend for 1963–2019 of  $-0.0167 (\pm 0.002).\text{decade}^{-1}$ . Most of the observed pH change in the Mozambique Channel is thus likely due to anthropogenic  $\text{CO}_2$ . This could impact not only the biological processes in the open ocean but also coral reef ecosystems in this region.

### 3.5. A Paleo-acidification perspective

Like in other regions, coral reefs in the Mozambique Channel are subject to multiple stressors due to warming, sea level rise and acidification, combined or not to local anthropization (e.g. in Mayotte). Historical SST and pH can be reconstructed from aragonite skeletal cores collected from massive scleractinian coral species like *Porites* sp. from geochemical proxies such as trace element ratios (e.g. Li/Mg, Sr/Ca), oxygen ( $\delta^{18}\text{O}$ ) or boron ( $\delta^{11}\text{B}$ ) isotopic ratio (e.g. Montagna et al., 2014; Tierney et al., 2015; Liu et al., 2014; Wu et al., 2018; D'Olivo et al., 2019; Cuny-Guirriec et al., 2019). Above we have estimated the change in pH since 1963. If we assume that the trends derived from observations between the sixties and 2019 are representative of the trend over the historical period (i.e.  $f\text{CO}_2$  tracks atmospheric  $\text{CO}_2$ ) historical changes in sea surface  $f\text{CO}_2$ , pH and  $N-C_T$  can be evaluated back to the pre-industrial period (Fig. 13a and b). We used historical atmospheric  $x\text{CO}_2$  data for the southern hemisphere reconstructed by Meinshausen et al. (2017) for the period 1800–2014 along with recent atmospheric  $x\text{CO}_2$  recorded over 2015–2019 (Dlugokencky and Tans, 2020).  $C_T$  and

**Table 7**

Mean  $C_{\text{ant}}$  accumulation and trends between 1994 and 2007 in the region 25°S/42°E in different layers (data from Gruber et al., 2019b). Standard-deviations are in bracket.

Layer	Delta- $C_{\text{ant}}$	Trend (1994–2007)
	$\mu\text{mol.kg}^{-1}$	$\mu\text{mol.kg}^{-1}.\text{yr}^{-1}$
0–30m	14.44 (0.05)	1.11 (0.004)
50–100m	14.04 (0.19)	1.08 (0.015)
100–150m	13.64 (0.15)	1.05 (0.012)
150–200m	12.94 (0.56)	1.00 (0.043)

pH were calculated assuming constant salinity (35) and alkalinity (2300  $\mu\text{mol.kg}^{-1}$ , the mean of all  $A_T$  observations in January 2004 and April 2019 was  $2297.6 \pm 14.7 \mu\text{mol.kg}^{-1}$ , Fig. 3a). We also assume that the oceanic  $f\text{CO}_2$  is either in equilibrium with the atmosphere (orange line in Fig. 13a and b) or that  $f\text{CO}_2$  seasonality is constant over time (grey lines in Fig. 13a and b).

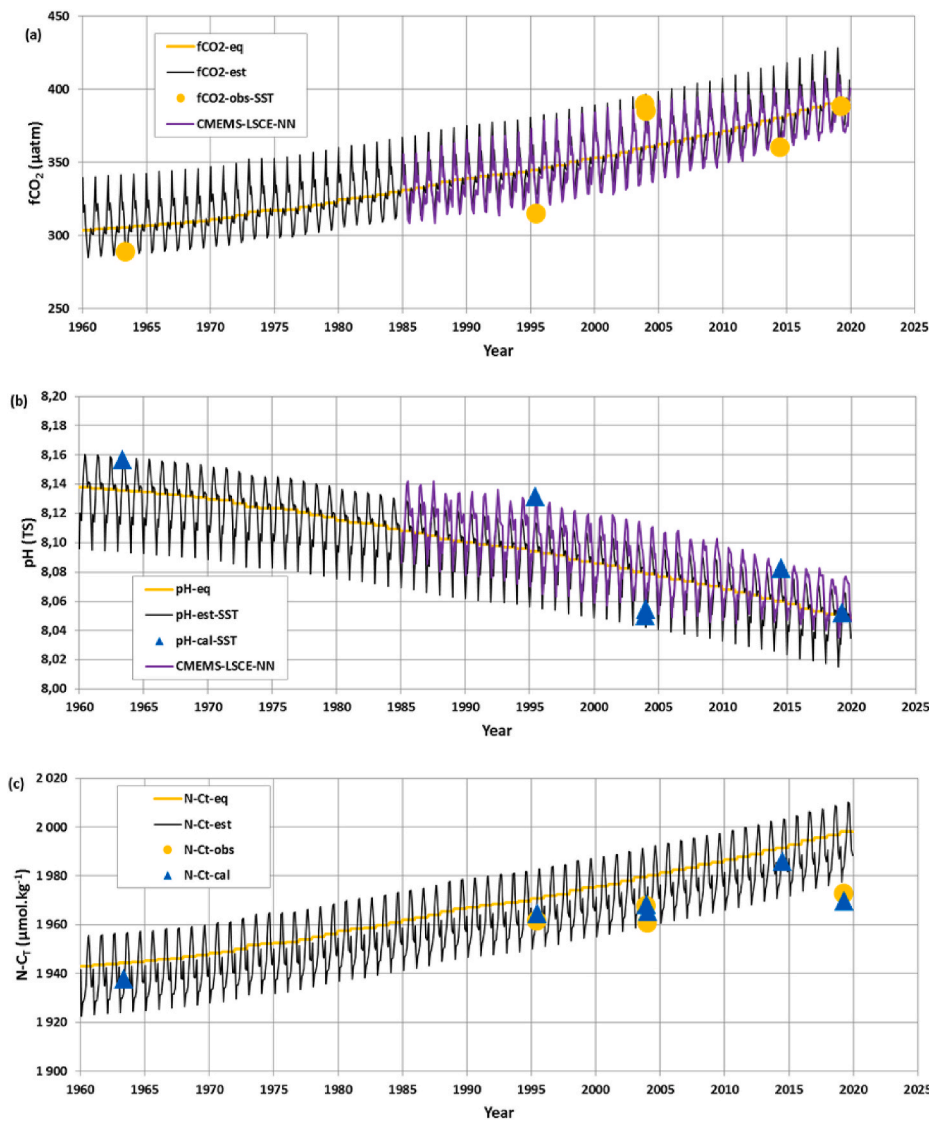
The results of the reconstruction for the recent decades (Fig. 13a) show a good agreement with observations for  $f\text{CO}_2$  and pH when available. For the period 1985–2019 the reconstructed  $f\text{CO}_2$  and pH are also close to the monthly values from the neural network model (Chau et al., 2020, purple lines in Fig. 13a). Reconstructed  $N-C_T$  concentrations are also in the range of  $N-C_T$  measured or calculated between 1963 and 2019. In the year 1800, values of  $f\text{CO}_2$ , pH and  $N-C_T$  were respectively around 270 ( $\pm 10$ )  $\mu\text{atm}$ , 8.18 ( $\pm 0.014$ ) and 1915 ( $\pm 10$ )  $\mu\text{mol.kg}^{-1}$ . This gives a reference for pH reconstructions based on 2 m-long coral cores recently collected at different locations along the Mozambique Channel during the CLIM-EPARSES cruise (work in progress, Tribollet, 2019, 2020).

Since 1800, the mean annual pH decreased progressively from 8.180 to 8.136 in 1963 (the first direct observation) and then decreased sharply to reach 8.05 in 2019. The pH decline of  $-0.13$  between 1800 and 2019, evaluated here at local scale, is slightly larger than the  $-0.11$  pH units estimated for the global ocean (Jiang et al., 2019). Based on our reconstruction, we evaluated a pH trend of  $-0.0027.\text{decade}^{-1}$  during 1800–1963,  $-0.0138.\text{decade}^{-1}$  during 1963–1995 and  $-0.0187.\text{decade}^{-1}$  during 1995–2019. Recalling our results based on observations (Table 5), it is clear that the rate of acidification has increased over the past recent decades, and it is likely that this process will continue in the next decade given the rapid increase of atmospheric  $\text{CO}_2$  concentrations that reached a new record of +2.6  $\text{ppm}.\text{yr}^{-1}$  in 2019 for a non ENSO year (Dlugokencky and Tans, 2020). If future atmospheric  $\text{CO}_2$  levels keep increasing at the same rate, a further decrease of pH by  $-0.1$  is likely to occur by 2040 ( $\text{pH} < 8$ ) while aragonite saturation state ( $\Omega_{\text{Ar}}$ ) would reach the value of 3 (Supp. Fig. S14) which is below the critical threshold of  $\Omega_{\text{Ar}} = 3.25$  that seems to limit the distribution of tropical coral reefs in the contemporary ocean (e.g. Hoegh-Guldberg et al., 2007).

Such a rapid reduction of pH and  $\Omega_{\text{Ar}}$  in the Mozambique Channel in response to anthropogenic  $\text{CO}_2$  emissions could have severe negative impacts on coral reefs because of a simultaneous reduction of calcification of the main reef framebuilders (corals and crustose coralline algae; Pandolfi et al., 2011) and a stimulation of carbonate dissolution processes (Schönberg et al., 2017; Eyre et al., 2018; Tribollet et al., 2019). Impacts on coral reefs of combined effects of ocean warming and acidification, together with local disturbances (e.g. eutrophication), need to be further investigated as the interaction of stressors might be synergistic or antagonistic (Chauvin et al., 2011; Trnovsky et al., 2016; Schönberg et al., 2017; Boyd et al., 2018). Future studies need to address impacts of multiple drivers on coral reefs at the regional scale, f.i. along thermal, pH and pollution gradients, to better apprehend and predict the fate of those ecosystems.

## 4. Summary and concluding remarks

This study presents new observations of the carbonate system in the Mozambique Channel for January 2004 and April 2019. Remarkable



**Fig. 13.** a: Reconstructed (black lines) sea surface  $f\text{CO}_2$  (a), pH (b) and  $\text{N-C}_T$  (c) for the periods 1960–2019 based on atmospheric  $x\text{CO}_2$  historical data. Also shown the mean results at  $25^\circ\text{S}$  based on observations (orange circles for  $f\text{CO}_2$  and  $\text{N-C}_T$ ) or calculated from  $f\text{CO}_2$  and  $A_T/\text{SSS}$  (blue triangles) as described in Fig. 10 and listed in Table 4. The monthly  $p\text{CO}_2$  and pH (at  $25^\circ\text{S}$ – $40^\circ\text{E}$ ) for 1985–2019 from the CMEMS-LSCE-NN model (Chau et al., 2020) is shown in purple line in (a) and (b). In all figures the orange curves are the reconstructed values assuming ocean  $f\text{CO}_2$  in equilibrium with the atmosphere. b: Same as Fig. 11a for the period 1800–2019. For clarity results from the CMEMS-LSCE-NN model for 1985–2019 not shown here.

differences are observed between these cruises conducted 15 years apart and at different seasons. The region was a large  $\text{CO}_2$  source in January due to high temperature ( $30^\circ\text{C}$ ) and near equilibrium in April. In 2019 this region was hit by the strong cyclone “Idai” leading to low salinity and low  $\text{C}_T$  and  $A_T$  concentrations. However, when normalized to salinity,  $\text{N-C}_T$  concentrations were higher in 2019 compared to 2004 by about  $12 (\pm 7) \mu\text{mol.kg}^{-1}$ , which reflects in part the uptake of anthropogenic  $\text{CO}_2$  over 15 years. Surprisingly the  $f\text{CO}_2$  (and pH) data at in-situ SST were almost identical in 2004 and 2019, although atmospheric  $x\text{CO}_2$  observed onboard increased by  $+34$  ppm. However, when the thermal effect is taken into account, we estimate a small increase in  $f\text{CO}_2$  ( $+11.5 \pm 9.8 \mu\text{atm}$ ) and decrease in pH ( $-0.012 \pm 0.008$ ) driven by  $\text{N-C}_T$  changes.  $\text{N-A}_T$  concentrations are the same in 2004 and 2019 and do not impact  $f\text{CO}_2$  and pH variations. The relatively small difference of  $f\text{CO}_2$  between the two cruises is due to seasonal variability that has to be taken into account to detect long-term trends. The decrease in  $\text{C}_T$  and  $f\text{CO}_2$  from January to April is in part due to biological processes. Diazotrophy provides a likely explanation as *Trichodesmium* spp has been observed in abundance in the Mozambique Channel (Dupuy et al., 2016). When the seasonality is taken into account, our observations in 2004 and 2019 reveal an increase in  $f\text{CO}_2$  of  $+1.75 (\pm 0.81) \mu\text{atm.yr}^{-1}$ , in  $\text{C}_T$  of  $+1.04 (\pm 0.79) \mu\text{mol.kg}^{-1}.\text{yr}^{-1}$  and a decrease of pH of  $-0.0016$ .

$\text{yr}^{-1} (\pm 0.0015)$ .

The data from these two cruises lead to a robust  $A_T/\text{SSS}$  relationship in this region that can be used to calculate pH from  $f\text{CO}_2$  data from other cruises. We attempted a comparison of our recent observations with sea surface data collected in 1963 in the Mozambique Channel (Keeling and Waterman, 1968). As expected, the  $f\text{CO}_2$  was much higher in 2004/2019 than in 1963 (by about  $100 \mu\text{atm}$ ) reflecting an atmospheric  $\text{CO}_2$  increase by  $+90$  ppm between 1963 and 2019. In addition to anthropogenic  $\text{CO}_2$  uptake, long-term ocean warming in this region ( $+0.11^\circ\text{C}.\text{decade}^{-1}$ ) contributes to  $f\text{CO}_2$  increase from 1963 to 2019 (by approximately about  $+10 \mu\text{atm}$ ). For pH, we observed a large decrease in all sectors of the Mozambique Channel, including near coral reef areas in the Eparses Islands (Europa, Juan de Nova). We estimated a mean decrease of  $-0.104 (\pm 0.006)$  pH unit between 1963 and 2019, typical of the preindustrial versus modern change in the global ocean (Jiang et al., 2019).

Results from the three cruises (1963, 2004 and 2019) covering a large region ( $14^\circ\text{S}$ – $25^\circ\text{S}$ ) are corroborated by three additional observations collected in the southern part of the Mozambique Channel ( $25^\circ\text{S}$ ). Based on those 6 cruises from 1963 to 2019, we evaluated a long-term trend for  $f\text{CO}_2$  of  $+1.55 (\pm 0.11) \mu\text{atm.yr}^{-1}$  almost identical to the atmospheric trend. For pH the trend over 1963–2019 is  $-0.00167.\text{yr}^{-1}$

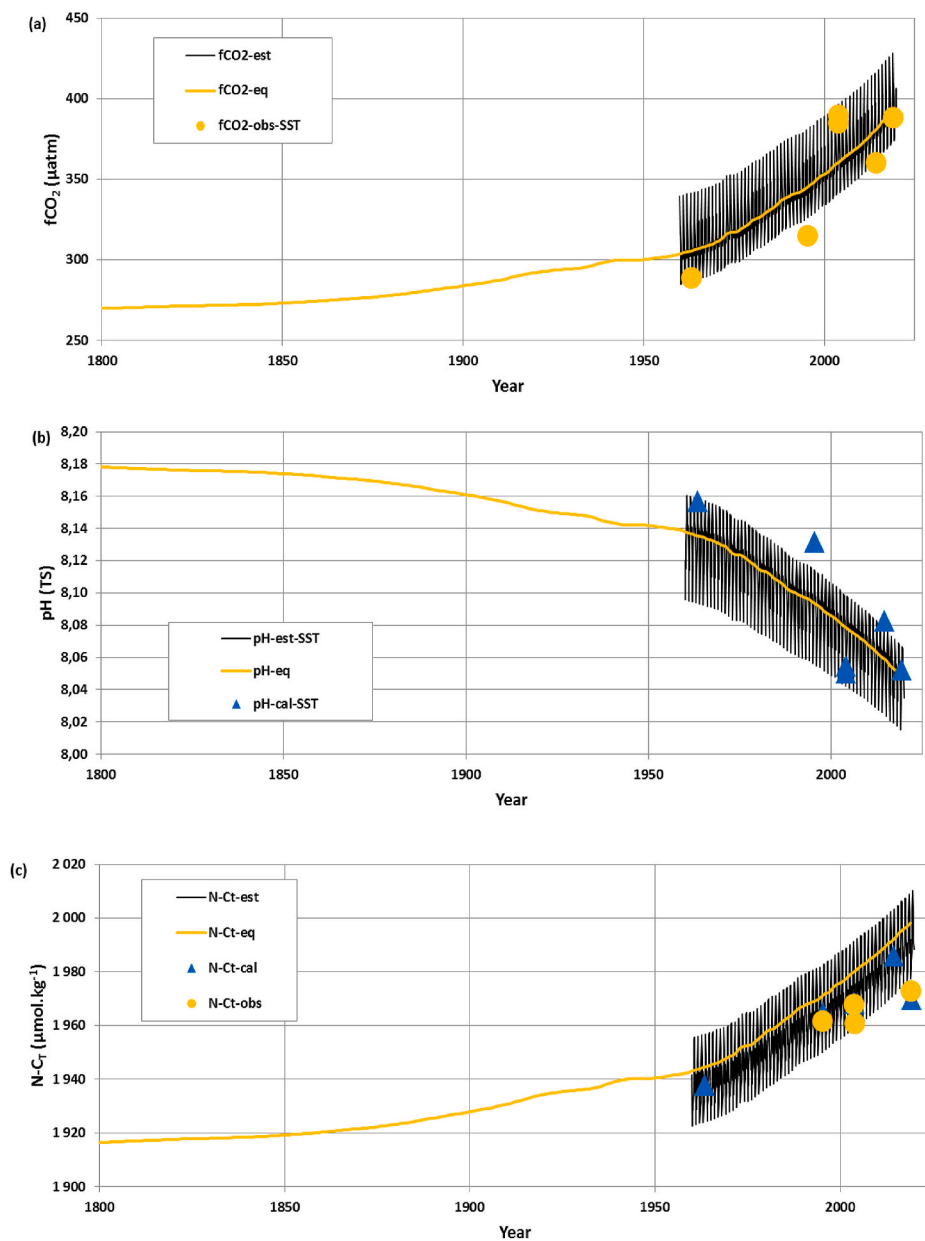


Fig. 13. (continued).

( $\pm 0.0002$ ), and for  $C_T$  we estimated an increase of  $+1.04 (\pm 0.21) \mu\text{mol.kg}^{-1}.\text{yr}^{-1}$ . This is close to the trend of anthropogenic  $\text{CO}_2$  evaluated in the upper ocean layers ranging between  $+0.93 (\pm 0.13)$  and  $+1.00 (\pm 0.04) \mu\text{mol.kg}^{-1}.\text{yr}^{-1}$  depending on the method and data used (Murata et al., 2010; Gruber et al., 2019a, this study). It is worth noting that the  $f\text{CO}_2$ , pH and  $C_T$  trends appear to be more pronounced in the recent decades although this is derived from few cruises only (Table 5). For pH we estimated a trend of  $-0.0129.\text{decade}^{-1} (\pm 0.0042)$  over 1963–1995 and  $-0.0227.\text{decade}^{-1} (\pm 0.0048)$  for 1995–2019 suggesting an acceleration of acidification. Our different estimates based either on  $f\text{CO}_2$  or  $A_T/C_T$  measurements in the Mozambique Channel agree with trends in this region derived from global scale reconstructions of  $p\text{CO}_2$  and pH (Denvil-Sommer et al., 2019; Chau et al., 2020). We also note that the pH trend in the Mozambique Channel appears lower than previous estimates at basin scale in the Indian Ocean which range between  $-0.024.\text{decade}^{-1}$  for the period 1981–2011 and  $-0.027.\text{decade}^{-1}$  for 1991–2011 (Lauvset et al., 2015).

The results presented in this analysis aimed at evaluating the change of the carbonate system in the Mozambique Channel at regional scale

and we conclude that the anthropogenic  $\text{CO}_2$  emissions are responsible for a significant acidification in surface waters supported here by various sea surface observations and independent anthropogenic  $\text{CO}_2$  concentration estimates in the water column. Given the results based on observations since the sixties, we reconstructed the pH change back to the pre-industrial period. In the year 1800, we estimated that pH in the Mozambique Channel was about  $8.18 (\pm 0.014)$ , i.e. 0.13 higher than in 2019. Concentration of  $C_T$  in the year 1800 was likely around  $1915 (\pm 10) \mu\text{mol.kg}^{-1}$ . These values could serve as a reference for reconstructing pH from coral core samples collected during the CLIM-EPARSES cruise in April 2019 (Tribollet, 2019, 2020).

Our analysis reflects only the change in the open ocean and the “remote” anthropogenic impact through  $\text{CO}_2$  emissions. To further evaluate impacts of global environmental change and ocean acidification on coral reefs, a continuous *in situ* sampling program will be needed at higher temporal and spatial scale. It should include the sea level changes either due to global warming or linked to marine heat waves or climate fluctuations such as the Indian Ocean Dipole (IOD) and ENSO in the Pacific (Ampou et al., 2017), the frequency of cyclone events or local

anthropization (especially in Mayotte). In the present analysis, pH was calculated from carbonate system properties, i.e. not from direct pH measurements. In the future and similarly to what is done in other coral reefs areas (Tilbrook et al., 2019), pH should be monitored at high frequency. Detailed analysis of the observations obtained during CLIM-EPARSEs cruise near the Islands and in coral reefs (Juan de Nova, Europa, Mayotte and Glorieuses) will be investigated in further studies.

#### Author's contribution

Claire Lo Monaco: Co-PI of the on-going OISO project, measured and qualified the  $fCO_2$ ,  $A_T$  and  $C_T$  data for OISO-11 and CLIM-EPARSEs cruises, contributed to the interpretation of the results and the writing of the manuscript. Nicolas Metzl is Co-PI of the on-going OISO project, was Chief scientist of the OISO-11 cruise, measured and qualified the  $fCO_2$ ,  $A_T$  and  $C_T$  data for OISO-11 cruise, qualified the  $fCO_2$ ,  $A_T$  and  $C_T$  data for CLIM-EPARSEs cruise, interpreted the results, wrote the draft of the manuscript and prepared the figures. Jonathan Fin measured the  $fCO_2$ ,  $A_T$  and  $C_T$  properties and helped for sampling during CLIM-EPARSEs cruise. Claude Mignon: measured the  $fCO_2$ ,  $A_T$  and  $C_T$  properties and helped for sampling during CLIM-EPARSEs cruise. Pascale Cuet: measured and qualified the Chl-a samples for CLIM-EPARSEs cruise, contributed to the interpretation of the results and the writing of the manuscript. Eric Douville: helped for the sampling during CLIM-EPARSEs cruise and contributed to the writing of the manuscript. Marion Gehlen: provided the neural network model results, contributed to the statistics, to the interpretation of the results and the writing of the manuscript. Trang Chau: provided the neural network model results, contributed to the statistics and prepared some figures and tables in the Supplementary material. Aline Tribollet: is PI of the ongoing CLIM-EPARSEs project, was Chief scientist of the CLIM-EPARSEs cruise and contributed to the interpretation of the results and the writing of the manuscript.

#### 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.dsr2.2021.104936>.

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