

## Wintertime process study of the North Brazil Current rings reveals the region as a larger sink for CO<sub>2</sub> than expected

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### Abstract

The key processes driving the air-sea CO<sub>2</sub> fluxes in the western tropical Atlantic (WTA) in winter are poorly known. WTA is a highly dynamic oceanic region, expected to have a dominant role on the variability of CO<sub>2</sub> air-sea fluxes. In early 2020 (February), this region was the site of a large in-situ survey and studied in wider context through satellite measurements. The North Brazil Current (NBC) flows northward along the coast of south America, retroflects close to 8°N and pinches off the world's largest eddies, the NBC rings. The rings are formed to the north of the Amazon River mouth when freshwater discharge is still significant in winter (a time period of relatively low runoff). We show that in February 2020, the region [50°W-59°W – 5°N-16°N] is a CO<sub>2</sub> sink from the atmosphere to the ocean (-1.7 TgC.month<sup>-1</sup>), a factor of 10 greater than previously estimated. The spatial distribution of CO<sub>2</sub> fugacity is strongly influenced by eddies south of 12°N. During the campaign, a nutrient rich freshwater plume from the Amazon River is entrained by a ring from the shelf up to 12°N leading to high phytoplankton concentration and significant carbon drawdown (~20 % of the total sink). In trapping equatorial waters, NBC rings are a small source of CO<sub>2</sub>. The less variable North Atlantic subtropical water extends from 12°N northward and represents ~60 % of the total sink due to their lower temperature associated with winter cooling and strong winds. Our results, in identifying the key processes influencing the air-sea CO<sub>2</sub> flux in the WTA, highlight the role of eddy interactions with the Amazon River plume. It sheds light on how lack of data impeded a correct assessment of the flux in the past, and on the necessity of taking into account features at meso and small scale.

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**a supprimé:** flux variability in winter 2020

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**a supprimé:** The North Brazil Current (NBC) flows northward across the Equator, passes the mouth of the Amazon River, and forms large oceanic eddies near 8°N. We investigate the processes driving the variability of air-sea CO<sub>2</sub> fluxes at different scales in early 2020 in the region [50°W-59°W – 5°N-16°N]. This region is a pathway between the equatorial and North Atlantic Ocean and was surveyed during the EUREC<sup>4</sup>A-OA/ATOMIC campaign. In-situ surface fugacity of CO<sub>2</sub> (fCO<sub>2</sub>), salinity and temperature combined with maps of satellite salinity, chlorophyll-a and temperature highlight contrasting properties in the region. In February 2020, the area is a CO<sub>2</sub> sink (-1.7 TgC.month<sup>-1</sup>), previously underestimated by a factor 10. The NBC rings transport saline and high fCO<sub>2</sub> water indicative of their equatorial origins and are a small source of CO<sub>2</sub> at regional scale. Their main impact on the variability of biogeochemical parameters is through the filaments they entrain into the open ocean. During the campaign, a nutrient rich freshwater plume from the Amazon River is entrained from the shelf up to 12°N and caused a phytoplankton bloom leading to a significant carbon drawdown (~20 % of the total sink). On the other hand, saltier filaments of shelf water rich in detrital material act as strong local sources of CO<sub>2</sub>. Spatial distribution of fCO<sub>2</sub> is therefore strongly influenced by ocean dynamics south of 12°N. The less variable North Atlantic subtropical water extends from Barbados northward. They represent ~60 % of the total sink due to their lower temperature associated with winter cooling and strong winds.

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## 60 1 Introduction

The North Brazil Current (NBC) is one of the dominant features of the tropical Atlantic circulation. In a region dominated by zonal jets, it flows northward along the coast of South America and separates from the coast around 6-8°N. The NBC seasonally turns back on itself in a tight loop, called the NBC retroflexion, and feeds the North Equatorial Counter Current, closing off the equatorial wind-driven gyre (Figure 1). This retroflexion occasionally pinches off some of the world's largest eddies, the North Brazil Current rings (Johns et al., 1990; Richardson et al., 1994).

After their separation from the NBC retroflexion region, the rings travel north-westward toward the Lesser Antilles in a course parallel to the coast of South America. These eddies have been extensively studied using modelling and both, in-situ (e.g. 1998-2001 NBC Ring experiment, Wilson et al., 2002) and satellite observations (e.g. Goni & Johns, 2001; Fratantoni & Glickson, 2002; Aroucha et al., 2020). They have a mean radius of 200 km and their diameter can exceed 450 km. Vertically, some of them extend down to more than 1000 m (Fratantoni and Glickson, 2002; Fratantoni and Richardson, 2006; Johns et al., 2003). The NBC rings swirl clockwise and travel with an average north-westward translation speed of 8-15 km d<sup>-1</sup> (Johns et al., 1990; Mélice and Arnault, 2017). Most of the anticyclonic eddies detectable by altimetry are rather shallow, extending from the surface to 200-300 m (Garraffo et al., 2003; Wilson et al., 2002). When they reach the Lesser Antilles, they start to coalesce and disintegrate, partly due to interactions with the topography (Fratantoni and Richardson, 2006; Jochumsen et al., 2010). There is substantial variability in the number of rings shed per year, ranging from 5 (Aroucha et al., 2020; Fratantoni and Glickson, 2002; Mélice and Arnault, 2017; Goni and Johns, 2001) to 9 (Johns et al., 2003). NBC rings play a crucial role in the interhemispheric transport of salt and heat in the Atlantic Ocean and are an important part of the meridional overturning circulation (Johns et al., 2003). The NBC rings disrupt an already complex region located in the vicinity of the Amazon River mouth and at the transition between equatorial and subtropical waters. While most of the studies on rings focused on their physical properties, little is known about their biogeochemical properties and how they affect the air-sea CO<sub>2</sub> flux of the western tropical Atlantic.

The global ocean acts as an atmospheric CO<sub>2</sub> sink, taking up 23% of total anthropogenic CO<sub>2</sub> emissions (Friedlingstein et al., 2020) and leading to ocean acidification (IPCC, 2021; 2019). The concentration of atmospheric CO<sub>2</sub> is increasing due to human activities (IPCC, 2019; 2021), and characterizing the role of the ocean in mitigating climate change through CO<sub>2</sub> uptake is thus a key investigation. The equatorial Atlantic Ocean is the second-largest source of CO<sub>2</sub> to the atmosphere after the equatorial Pacific (Landschützer et al., 2014; Takahashi et al., 2009). Previous works in this region examined the influence of the equatorial upwelling and of the Amazon plume on the CO<sub>2</sub> flux. CO<sub>2</sub>-rich equatorial waters, originating from the equatorial upwelling (Andrié et al., 1986) strongly contrast with the CO<sub>2</sub>-undersaturated Amazon River plume waters. The magnitude of the Amazon River discharge is unique in the global ocean. It represents as much freshwater as the next 7 largest rivers in the world combined and contributes to almost 20% of global river freshwater input to the ocean (Dai and Trenberth, 2002). It

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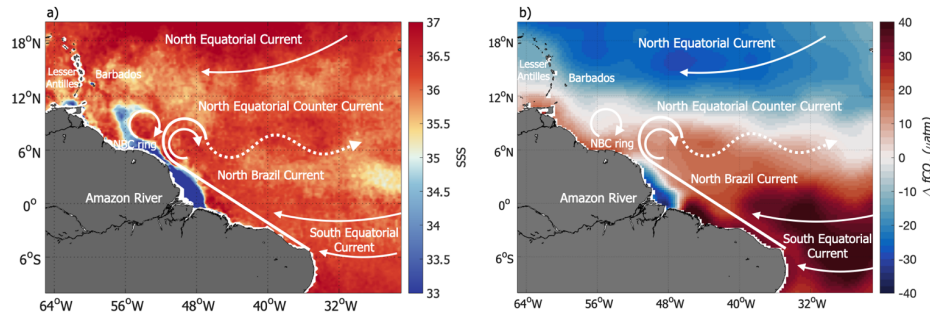
150 therefore strongly impacts the physical, biogeochemical and biological properties of the coastal and the open ocean. Often overlooked, the Amazon River plume is an atmospheric CO<sub>2</sub> sink of global importance (Ibáñez et al., 2016). The plume carries water rich in silicate, nitrogen and phosphate into the tropical oceanic waters that are strongly depleted in nutrients. As water mixes and turbidity decreases, the primary producer's growth and associated biological drawdown are stimulated (Chen et al., 2012). Nitrogen is rapidly consumed, and nitrogen fixation by diazotrophs becomes the main pathway of carbon sequestration in the plume (Subramaniam et al., 2008). This strong carbon drawdown leads to a significant sink of atmospheric CO<sub>2</sub> (Körtzinger, 2003; Lefèvre et al., 2010). Not taking into account the Amazon plume would result in overestimating the tropical Atlantic air-sea CO<sub>2</sub> flux by 10% (Ibáñez et al., 2016).

155 The Amazon River's discharge reaches a minimum in December and progressively increases from January onwards. The plume extension is minimum from January to March (Fournier et al., 2015) and as a result, it is the period of maximum salinity in the northwestern tropical Atlantic. The Amazon outflow region is particularly hard to reconstruct due to its strong variability and a severe lack of data. Waters located in the southeasternmost part of the domain act as a strong source of CO<sub>2</sub> to the atmosphere. The source gradually turns into a sink north of 10°N as waters become colder due to seasonal winter cooling. This situation is typical of a transition zone between equatorial and subtropical waters in winter (Landschützer et al., 2020, Figure 1b). The northwestern tropical Atlantic is commonly divided into two parts, the northern much less variable part (also called "Trade wind region"), and the southern part, also referred as Eddy Boulevard (Stevens et al., 2021). The freshwater of the Amazon remains mainly confined to the continental shelf due to winds perpendicular to the coast as it travels northward into the Caribbean Sea (Coles et al., 2013). However, it has recently been documented that off-shore freshwater transport is often present in February and significantly alters the physical properties of the region (Reverdin et al., 2021). This is partly due to the interaction of the NBC rings with the Amazon plume (Figure 1a). The ocean color signature of the Amazon (Muller-Karger et al., 1988) has been used as a tracer to delineate the rings, and better understand their generation, evolution and characteristics (Johns et al., 1990; Fratantoni & Glickson, 2002). The Amazon River also influences the surface temperature and salinity of the rings. For example, Figure 1a shows a freshwater plume stirred by a large ring. They are considered warm-core rings but have a warm SST anomaly in the first half of the year, and a cold one in the second half, because the anomaly is relative to the regional SST, with an extensive warm pool in late summer and autumn (Ffield, 2005). Their signature in salinity is therefore plume-dependent as well. Ffield (2005) reported that 3 out of 4 rings were surrounded by lower salinity water. Salinity and chlorophyll-a are therefore critical to understand the surface physical and biogeochemical properties of the region, as well as the air-sea fluxes of CO<sub>2</sub>.

175 The northwestern tropical Atlantic is a dynamically active region, with eddies several hundred kilometers in diameter and connected to the world's largest river. There are surprisingly few biogeochemical observations available for winter months during low outflow of the Amazon River. Few tropical Atlantic measurements of biogeochemical tracers are available with one transect in winter in the Surface Ocean CO<sub>2</sub> Atlas (SOCAT, Bakker et al., 2016) database south of 10°N crossing the

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region. This scarcity is a major impediment in understanding the biological and physical processes underlying the oceanic carbon and nutrient cycles in the region. Satellite salinity shows a contrasted spatial structure, with eddies and filaments (Figure 1a). In this study, we take advantage of the physical and biogeochemical data collected during the EUREC<sup>4</sup>A-OA/ATOMIC experiment in January-February 2020, combined with satellite data, to understand how the NBC rings and their related structures impact the air-sea CO<sub>2</sub> flux in winter. The paper is organized as follows. We present the in-situ observational data from the EUREC<sup>4</sup>A-OA/ATOMIC experiment as well as the satellite data in section 2. We identify the water masses observed in the region, their physical and biogeochemical properties and estimate the CO<sub>2</sub> fluxes at regional scale using empirical relationships in section 3. In section 4, we compare the results with climatologies of air-sea CO<sub>2</sub> fluxes to evaluate the added knowledge brought by the intensive surveys of February 2020. We discuss the results and the interannual variability in section 4 and we conclude in section 5.



**Figure 1:** Schematic of the main ocean currents in the western tropical Atlantic superimposed over the SSS field of Feb. 7<sup>th</sup> 2017 (a) and over the February ΔfCO<sub>2</sub> climatology from Landschützer et al., 2020 (b).

## 2 Data and Methods

### 2.1 In-situ data

The EUREC<sup>4</sup>A-OA (Elucidating the Role of Clouds-Circulation Coupling in Climate Ocean-Atmosphere) / ATOMIC (Atlantic Tradewind Ocean Mesoscale Interaction Campaign, Stevens et al., 2021) campaign took place in January and February 2020 and involved research vessels (RV) from France (RV Atalante, Speich and The Embarked Science Team, 2021), Germany (RV Maria S. Merian, hereby designated as Merian, Karstensen et al., 2020 and RV Meteor, not considered in this

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280 study since no CO<sub>2</sub> measurements were taken onboard), and the United States (RV Ronald H. Brown, hereby designated as Ron Brown, Quinn et al. 2021). These cruises provided numerous in-situ measurements and, in this study, we will focus on the continuous near surface measurements of temperature, salinity and fCO<sub>2</sub>.

285 Temperature, and salinity from thermosalinographs (TSG), as well as fCO<sub>2</sub>, were measured from water pumped ~5 m below the surface. For each ship, the resulting CO<sub>2</sub> data is corrected (Lefèvre et al., 2010; Pierrot et al., 2009) from the temperature difference between the water at the ship's water intake and the one analyzed by the instrument. The RV Atalante fCO<sub>2</sub> measurements started on January 30<sup>th</sup> and ended on February 18<sup>th</sup> 2020 (Olivier et al., 2020). The underway oceanic and atmospheric fCO<sub>2</sub> were detected by infrared detection using a Licor 7000 (Takahashi et al., 1993). The fCO<sub>2</sub> system was the same as in Lefèvre et al. (2010). It uses a shower air-sea equilibrator described by Poisson et al. (1993). Seawater from the  
290 TSG pumping circuit circulates in the equilibrator at a rate of 2 L.min<sup>-1</sup>. A closed-loop of about 100 mL of air flows through the equilibrator designed to avoid bubbles at the air-sea interface. To minimize temperature corrections, the equilibrator is thermostated with the same seawater as the one used for CO<sub>2</sub> measurements. The temperature difference between the equilibrator and the sea was on the order of 0.5 °C.

295 Furthermore, 138 samples for dissolved inorganic carbon (DIC) and total alkalinity (TA) analysis were collected onboard RV Atalante as well as inorganic nutrients (silicate, phosphate, nitrate and nitrite). DIC and TA were measured at the SNAPO-CO<sub>2</sub> facility by potentiometric titration using a closed cell, following the method of Edmond (1970). Nutrients were conserved by heat pasteurization and analysed by colourimetry at IRD LAMA service in Brest.

An OceanPackCUBE ferrybox system from SubCtech was installed on the RV Merian measuring continuously the oceanic fCO<sub>2</sub> from the 23<sup>rd</sup> of January to the 19<sup>th</sup> of February 2020. Water is pumped at a rate of ~7 L.min<sup>-1</sup> through a debubbler unit  
300 subsequently followed by a SeaBird SBE 45 thermosalinograph before it circulates along a membrane through which CO<sub>2</sub> diffuses. On the other side of the membrane, the air-loop is circulated at a rate of 0.5 L.min<sup>-1</sup> through a Li-COR LI840 non-dispersive infrared gas analyzer (e.g., Arruda et al., 2020). The RV Ron Brown completed two legs, from January 10<sup>th</sup> to January 25<sup>th</sup> and from January 29<sup>th</sup> to February 13<sup>th</sup> 2020. The General Oceanic Inc. 8500 pCO<sub>2</sub> instrument installed onboard, follows a similar methodology to the underway fCO<sub>2</sub> system deployed on the Atalante and is detailed in Pierrot et al. (2009).

305 An intercomparison of the fCO<sub>2</sub> measured by the RVs Atalante and Merian is attempted when the ships were navigating together at a distance less than 5 km (Figure 2). On average, the fCO<sub>2</sub> measured by the RV Merian is 6.4 µatm higher than the one measured on the RV Atalante, with a standard deviation of 4.8 µatm. The RVs Merian and Ron Brown crossed the same water mass at 13-14°N/57°W on February 12<sup>th</sup>. On average, the RV Merian fCO<sub>2</sub> is 6 µatm higher than the RV Ron Brown  
310 fCO<sub>2</sub>. In part we link these differences to the slower response time of the membrane system, however differences also lie within the uncertainties of the fCO<sub>2</sub> observing systems (~5 µatm for the membrane system installed on the RV Merian (see Arruda et al., 2020) and ~2 µatm for the equilibrator systems installed on the RVs Atalante and Ron Brown). The region where

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RV Merian and RV Atalante were close is very variable (standard deviation of  $20 \mu\text{atm}$ ), and the RV Merian and RV Ron Brown were never in the same place at the same time, so that the observed differences could also be due to the natural variability of  $f\text{CO}_2$  sampled differently by the various ships. Hence, we did not apply any correction and we checked that the effect of a  $6 \mu\text{atm}$  systematic bias on the RV Merian  $f\text{CO}_2$  has a minor effect on our resulting interpolations. A comparison between the reconstructed flux with and without a correction for the  $6 \mu\text{atm}$  systematic bias suggests that such a systematic bias would lead to less than  $2 \mu\text{atm}$  difference on our mean interpolated  $f\text{CO}_2$  and less than  $0.1 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$  on the mean derived air-sea  $\text{CO}_2$  flux.

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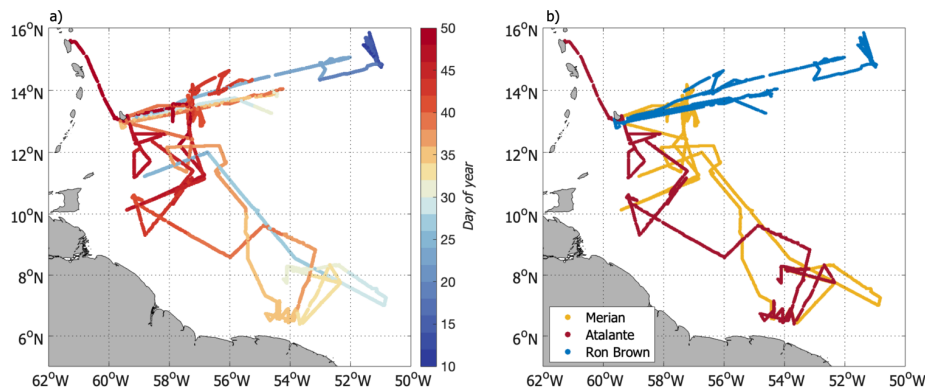


Figure 2: Ship tracks colour-coded by day of year (a) and by ship name (b).

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## 2.2 Satellite and atmospheric reanalyzes data

Daily satellite maps of chlorophyll-a (Chl $a$ ), sea surface temperature (SST), as well as absolute dynamic topography (ADT) and sea surface salinity (SSS) are used in this study.

The salinity maps are a blend of the Soil Moisture Ocean Salinity (SMOS, Jan. 2010-present), and Soil Moisture Active Passive (SMAP, Apr. 2015-present) measurements developed by Reverdin et al. (2021). The European SMOS and US SMAP missions observe the sea surface by L-band radiometry from sun-synchronous polar-orbiting satellites (Entekhabi et al., 2010; Font et al., 2009; Kerr et al., 2010; Piepmeier et al., 2017). Combining 6 a.m. and 6 p.m. measurements of both missions provides an

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almost complete coverage each day. When the coverage was not complete over our region, the 6 a.m. track of the following day was also included. This daily field is available for the first 20 days of February, and leaves out only 2 days without sufficient coverage to retrieve salinity data. It has a spatial resolution close to 70 km, and an uncertainty on the order of 0.5.

This product is optimized for the northwestern tropical Atlantic in February 2020 and has an almost daily resolution. It is an experimental daily product built to have the best representation possible of the Amazon plume variability. The product, its uncertainties and the comparison between the TSG salinity and the satellite SSS are detailed in Reverdin et al. (2021).

Daily Chl<sub>a</sub> concentration maps and SST maps are produced by CLS (Stum et al., 2016) on a spatial grid of 0.02°. The Chl<sub>a</sub> concentration maps are composites built from VIIRS (on Suomi-NPP and NOAA-20 US platforms) and OLCI (on Sentinel 3A and 3B Copernicus European platforms) satellite sensors. The SST product is a 1-day average of 4 infrared radiometer satellite data. Both datasets are sensitive to the cloud cover, but during our period of interest, they are usually without many gaps except at the end of February. Comparison between the TSG SST and the satellite SST product are detailed in the RV Atalante cruise report (Speich & The Embarked Science Team, 2021).

Daily ADT maps at a ¼° resolution combine data from all satellites available for the period 1993 to present. From these ADT fields, the TOEddies algorithm, developed by Laxenaire et al. (2018), identifies eddies and their trajectories. The eddy detection is based on the closed contours of ADT, as well as the maximum geostrophic velocity associated to the eddy.

In order to compute the air-sea CO<sub>2</sub> fluxes, the European Centre for Medium-Range Weather Forecasts (ECMWF) Reanalysis v5 (ERA5) hourly wind speed and mean sea level pressure, P<sub>atm</sub>, is used. ERA5 covers the period from January 1950 to present, and provides hourly data on a 30 km grid. In addition, the monthly wind speed and SST fields over the period 1998-2015 are used, and a climatology over this period is computed. The wind speed in the region in winter is on average between 6 and 8 m s<sup>-1</sup> and its variability is low.

We compare the EUREC<sup>4</sup>A-OA/ATOMIC observations with the observation-based CO<sub>2</sub> partial pressure (pCO<sub>2</sub>) climatology developed by Landschützer et al., (2020), created using a 2-step neural network method (Landschützer et al., 2016) and combining open and coastal ocean datasets. The associated ΔpCO<sub>2</sub> and air-sea CO<sub>2</sub> flux monthly field climatologies over the 1998-2015 period are computed using the ERA5 climatological wind, SST and P<sub>atm</sub> fields as well as the atmospheric CO<sub>2</sub> from the Ragged Point, Barbados station.

## 2.3 Methods

### 2.3.1 Air-sea CO<sub>2</sub> flux

We compute the air-sea flux (F; mmol.m<sup>-2</sup>.day<sup>-1</sup>) as:

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a supprimé: From these ADT maps, the TOEddies algorithm, developed by Laxenaire et al. (2018), identifies eddies and their trajectories (Figure 1).

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$$F = k \cdot K_0 \cdot (fCO_{2a} - fCO_{2atm}) \quad (1)$$

420 Where  $K_0$  is the solubility of  $CO_2$  in seawater, expressed as a function of SSS and SST by Weiss (1974);  $fCO_{2atm}$  is the atmospheric  $CO_2$  fugacity; and  $k$  is the gas transfer velocity.  $k$  is calculated following the relation from Wanninkhof (2014);

$$k = 0.251 \cdot \langle U_{10}^2 \rangle \cdot (Sc/660)^{-0.5} \quad (2)$$

where  $Sc$  is the Schmidt number and  $U$  is the wind speed at 10 m above sea level derived from ERA5 wind speed. The ERA5 wind speed is used for the satellite-based analysis and the air-sea  $CO_2$  flux climatology. The measured winds from the ships are adjusted to 10 m following a logarithmic profile (Tennekes, 1973) and used to compute the along-track flux for visualisation purposes.

425 In order to compute  $fCO_{2atm}$  over that period, we first derived the saturation vapor pressure ( $P_{H_2O}$ ) from SSS and SST then the atmospheric  $pCO_2$  using the monthly averaged  $CO_2$  mole fraction ( $xCO_{2atm}$ ) measured at the NOAA/Earth System Research Laboratory (ESRL) station in Ragged Point, Barbados (13.17°N, 59.43°W):

$$430 fCO_{2atm} = xCO_{2atm} \cdot (P_{atm} - P_{H_2O}) \cdot C_f \quad (3)$$

Where  $C_f$  is the fugacity coefficient, function of the atmospheric pressure and SST (Weiss, 1974).

### 2.3.2 Reconstruction of $fCO_2$ from satellite maps

435 Our approach is to derive from the large EUREC<sup>4</sup>A-OA dataset a relationship linking  $fCO_2$  to SST, SSS and  $Chl_a$  in order to provide maps of  $fCO_2$  based on the satellite maps of SST, SSS and  $Chl_a$ .  $Chl_a$  was not measured onboard, thus, we use satellite surface  $Chl_a$  co-located along the ship track. This set of parameters is used as proxy to describe the influence of ocean dynamics, chemistry, and also marine biology on  $fCO_2$ . The biological carbon pump is one of the major components of the oceanic and global carbon cycles, as the photosynthetic production of organic carbon by marine phytoplankton accounts for about half of the carbon fixation associated with global primary production (Arrigo, 2007; Behrenfeld et al., 2006; Field et al., 1998). However, while  $Chl_a$  is an indicator of biological activity, it is also a very good tracer of ocean circulation so that, depending on the water masses origin,  $fCO_2$  and  $Chl_a$  are not expected to be systematically negatively correlated. Waters rich in detrital material tends to limit the phytoplankton growth and microbial respiration of riverine material on the continental shelf likely dominates (Aller and Blair, 2006; Medeiros et al., 2015; Mu et al., 2021). Even considering the extend of the EUREC<sup>4</sup>A-OA atomic cruise, the dataset is still sparse, and cannot fully represent the small-scale variability it highlights. In order to understand the fluxes at regional scale the need for a good spatial resolution arises. For that, the surface T-S- $Chl_a$  diagram computed from the ship measurement (and collocated satellite  $Chl_a$ ) is interpolated using a linear 3D interpolation on a grid of SST, SSS and  $Chl_a$ . The grid has a resolution of 0.01°C in SST, 0.1 in SSS, and 0.01 in  $\log(Chl_a \text{ (mg m}^{-3}\text{)})$ . Using a 3D linear interpolation to mapping the  $fCO_2$  data over a grid is a simple yet effective solution for a dataset that is still relatively sparse. The method is presented in more details in the supplementary materials (Text S1). Using a linear fit prevents oscillations between two data points, and yields good results. Along the ship track, the standard deviation between the measured and

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- a supprimé: where  $Sc$  is the Schmidt number and  $U$  is the wind speed at 10 m above sea level measured by each ships weather station or derived from ERA5 wind speed. The measured winds from the ships are used to compute the along-track flux, while the ERA5 wind speed is used for the satellite-based analysis and the air-sea  $CO_2$  flux climatology.<sup>4</sup>
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reconstructed  $fCO_2$  is  $4 \mu atm$ . To each triplet of surface T, S, and  $\log(Chla)$  in the range of the values measured by the ship is therefore associated a value of  $fCO_2$  based on the 3D linear interpolation of the in-situ values. In order to cover the whole range of T-S- $\log(Chla)$  present in the region, we extrapolate to lower temperatures and lower salinities than the ones measured by the ship. In order to do so, we add 4 points to the T-S- $Chla$  diagram at lower salinities and lower temperatures based on previous knowledge of the region. For the low salinity domain ( $SSS < 30$ ),  $fCO_2$  is strongly dominated by salinity and the influence of temperature is weak (Lefèvre et al., 2010). The SSS- $fCO_2$  relation developed by Lefèvre et al. (2010) is in good agreement with the SSS- $fCO_2$  relationship computed from this study data (supplementary Figure S1) in the common range, we therefore use it to compute  $fCO_2$  at a salinity of 26 ( $fCO_2(S = 26) = 251.4 \mu atm$ ). The lower temperature is mostly located in the northern part of the domain, that is the least variable and where the variations of  $fCO_2$  are dominated by the ones in temperature. From this dataset we compute a variation of  $15 \mu atm/^\circ C$ , which is consistent with the  $4.23\% \text{ } ^\circ C^{-1}$  expected variation of  $fCO_2$  with temperature due to the temperature sensitivity of the carbonate dissociation constants and  $CO_2$  solubility (Takahashi et al., 2002; Wanninkhof et al., 1999). We use this dependency to compute the  $fCO_2$  at a temperature of  $24^\circ C$  to cover the whole range of temperature in the region.

We combine the interpolated  $fCO_2$  with satellite maps of SST, SSS and  $Chla$  to obtain daily high-resolution maps of  $fCO_2$ . Some days, either the presence of clouds altering the  $Chla$  and SST and the lack of salinity coverage prevent the retrieval of  $fCO_2$ . In order to limit the error on  $fCO_2$ , we only keep 9 days out of the 20 first days of February (2, 4, 6, 7, 9, 11, 12, 17 and 19 of February) where the coverage is sufficient. Then, daily mean sea level pressure maps and wind fields are used to compute the air-sea  $CO_2$  flux over the region in a similar way as described in 2.3.1. The salinity maps present major errors near islands, because no correction of the island effect was applied on the SMAP maps (Grotsky et al., 2018). Therefore, the reconstructed flux will be studied over a region excluding the close vicinity of the islands [ $59^\circ W$ - $50^\circ W$ ,  $5^\circ N$ - $16^\circ N$ ].

### 3 Results

#### 3.1 A transition region presenting a strong mesoscale activity

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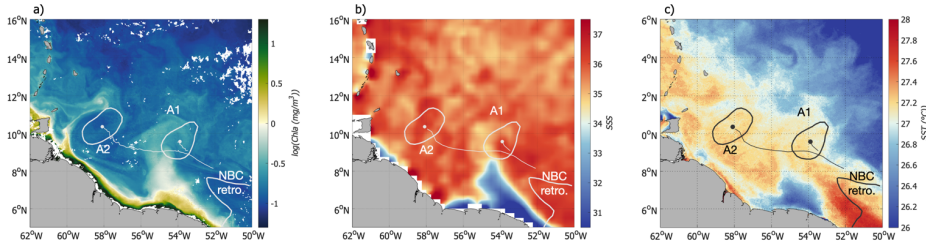
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**Figure 3: a) Chlorophyll-a, b) SSS and c) SST on Feb 6<sup>th</sup> 2020 with the contours of NBC rings A1 and A2, their centre and their trajectory. The NBC retroflexion is identified from the 0.51 m contour of the satellite derived ADT.**

Figure 3 shows how in February 2020, the ocean currents of the WTA strongly influence the variability of SSS, SST and Chl<sub>a</sub> at many scales, with an NBC ring stirring a plume of fresher water rich in chlorophyll-a toward the open ocean. This is also supported by the measurements done during the EUREC<sup>4</sup>A/ATOMIC campaign in January-February 2020 (Figure 4). They show a complex environment, for example  $\Delta fCO_2$  presents similar large-scale features as the climatology, but it also reveals numerous smaller scale structures (Figure 4a). Among the latter, two stand out. These are the very low  $fCO_2$  in the south-eastern part of the domain, and the high  $fCO_2$  around 11°N. In early 2020, the NBC retroflexion was very variable and shed two large anticyclonic rings (Figure 3). They are long-lived 250-km large eddies traveling north-westward toward the Lesser Antilles in the Eddy Boulevard region. The ring detection algorithm TOEddies based on ADT indicates that NBC ring A2 separated from the retroflexion in late December, was fairly stationary during the cruise period (February 2020) and located around 58°W-11°N. NBC ring A1 separated from the retroflexion in early February 2020 and then stayed around 54°W-10°N for 10 days before translating northwestward toward the Lesser Antilles after the 20<sup>th</sup> of February. These eddies contribute to the variability of the region in two ways. As they travel, they transport the water trapped in their core during their formation (eddy trapping), but they also stir the surrounding waters (eddy stirring).

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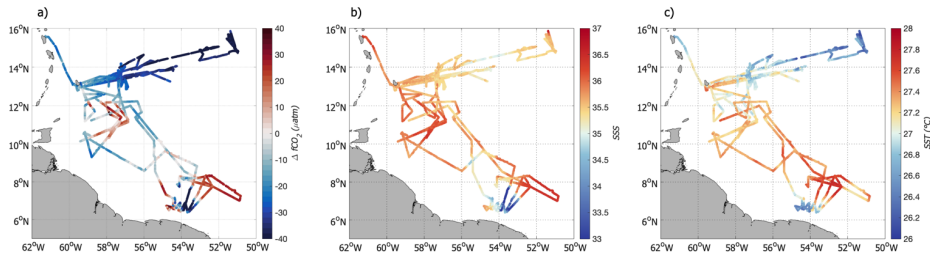
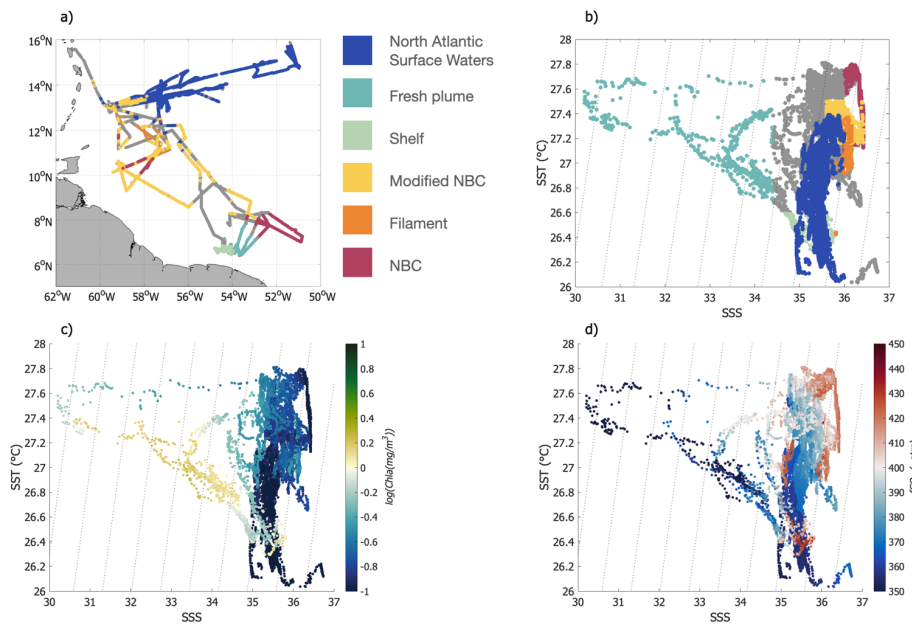


Figure 4: In-situ measurements of (a)  $\Delta f\text{CO}_2$ , (b) salinity, (c) temperature.

### 3.2 Surface water masses identification



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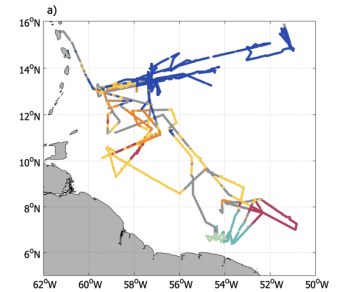
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720 **Figure 3:** a) Map representing the RVs Atalante, Merian and Ron Brown ship tracks, colour-coded with the identified water masses. b) T-S diagram, colour-coded with the water masses; the grey colour corresponds to points that do not fit into the definition of the identified water masses.

725 In an effort to understand how biogeochemistry is forced by physical processes in the ocean, we used surface Chl<sub>a</sub> to complement SST and SSS data in defining surface water masses. We observed that in the northwestern tropical Atlantic in winter in situ fCO<sub>2</sub> strongly depends on these three variables (Figure 4, Figure 5). There is a strong positive dependence of fCO<sub>2</sub> on SSS, with low fCO<sub>2</sub> for low SSS (Figure 5c-d). Across the whole EUREC<sup>4</sup>A-OA/ATOMIC region, SST did not vary much (mean SST of 27°C, and standard deviation of 0.5°C), but warmer waters present higher fCO<sub>2</sub>. The dependence on Chl<sub>a</sub> allows for the discrimination of water masses with the same surface TS properties but not the same fCO<sub>2</sub> (Figure 5). Satellite-based Chl<sub>a</sub> is hard to discriminate from detrital material using ocean colour where both are present as they have close spectral characteristics. Figure 5 shows that waters with an SST of 26.5°C and SSS between 35-36 can either be rich in Chl<sub>a</sub> and have a high fCO<sub>2</sub> or low in Chl<sub>a</sub> and have a low fCO<sub>2</sub>. By combining SST and SSS with Chl<sub>a</sub> and using information from the dynamical structures of the region, we identified six upper-ocean water masses (Figure 5a-b). The along track ΔfCO<sub>2</sub> for each ship is presented on Figure 6, colour-coded with the identified water mass, highlighting the link between the surface T-S-Chl<sub>a</sub> relation and ΔfCO<sub>2</sub>. The way we defined water masses, considering time-varying boundaries, is relatively similar to the one used by Longhurst (2010), and some of the surface water masses compare well with Longhurst (2010) biogeochemical provinces. He identified 3 provinces in the Northwestern tropical Atlantic, the North Atlantic Tropical Gyre province (NATR), the Western Tropical Atlantic province (WTRA) and the Guianas Coastal province. In this first part, we will present two surface water masses that are usually identified in the region (e.g. Longhurst et al., 1995, 2010) and their physical properties. North of Barbados, the domain is mostly dominated by North Atlantic Subtropical Waters (NASW), similar to Longhurst's NATR. They have a SSS in the range of 35 to 36 and are relatively cold (Table 1). Their SST diminishes over time towards the end of February. These waters are less influenced by coastal dynamics and therefore are not very productive at the surface (Chl<sub>a</sub> levels inferior to 0.14 mg.m<sup>-3</sup>) due to low nutrient levels. They are mainly located north of 13°N, and get progressively colder toward the north-east. The RV Ron Brown stayed in that Trade Wind region for almost a month (Figure 5). The observations collected from this ship show NASW lower fCO<sub>2</sub> with respect to the atmosphere (ΔfCO<sub>2</sub> = -40 μatm). Similar results are found on one of the RV Merian's transect, with ΔfCO<sub>2</sub> < -30 μatm.

740 The NBC is surface-intensified and fed by the central branch of the South Equatorial Current (SEC, Schott et al., 1998). As the cold and saline water from the upwelling region is transported westward by the SEC, it warms up (SST > 27°C), but retains its saline characteristic (SSS > 36, Table 1) as it reaches the NBC retroflection region (Figure 4). This NBC water mass is oligotrophic (Figure 3b), and therefore in our area of interest distinguished itself by its low level of surface Chl<sub>a</sub> (Chl<sub>a</sub> < 0.14 mg.m<sup>-3</sup>). These waters are found in the retroflection area, and sampled by both RVs Merian and Atalante at the beginning of

745 February (Figure 5). The NBC water mass is in some way similar to the WTRA, but does not extend beyond the retroflection,

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a supprimé: In order to better understand the variability of the region, it is necessary to characterize the main water masses present in winter. From SST and SSS only, we are not able to define water masses with a unique fCO<sub>2</sub> signature (Figure 3).

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therefore representing a more limited part of the WTRA. We will introduce four new water masses in the following parts, as well as their associated dynamical structures. They can be considered as subsets of the WTRA and Guiana Coastal province, these two provinces being too large and not well suited to represent small-scale processes.

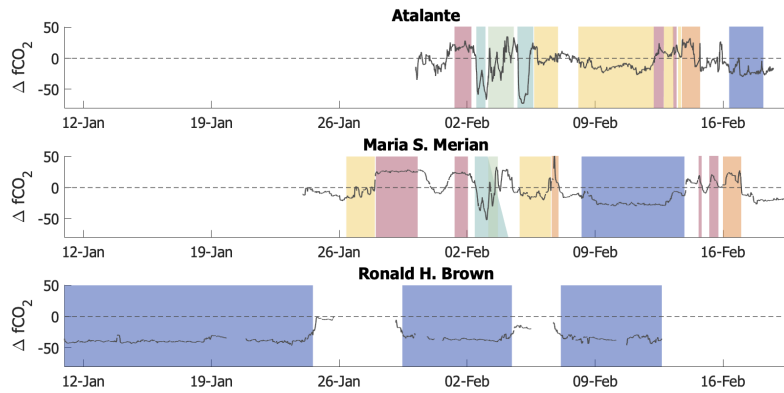


Figure 4: RVs Atalante (top), Merian (middle) and Ron Brown (bottom)  $\Delta f\text{CO}_2$  time-series. The background color indicates the crossed water mass domains (see definition in legend of Fig 5).

	<u>NASW</u>	<u>NBC</u>	<u>Modified NBC</u>	<u>Freshplume</u>	<u>Shelf</u>	<u>Filament</u>
<u>Temperature</u> (°C)	<u>&lt;27.2</u>	<u>&gt; 27</u>	<u>27.16 &lt; SST &lt; 27.6</u>		<u>&lt; 26.6</u>	<u>&lt; 27.4</u>
<u>Salinity</u>	<u>35 &lt; SSS &lt; 36</u>	<u>&gt; 36</u>	<u>&gt; 35.6</u>	<u>&lt; 34.5</u>		<u>35.8 &lt; SSS &lt; 36.3</u>
<u>Chlorophyll-a</u> (mg.m <sup>-3</sup> )	<u>&lt; 0.14</u>	<u>&lt; 0.14</u>	<u>0.11 &lt; Chla &lt; 0.25</u>	<u>&gt; 0.25</u>	<u>&gt; 0.25</u>	<u>&gt; 0.25</u>

Table 1. Thresholds in SSS, SST and Chla used to define the 6 water masses identified in the winter WTA.

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### 3.3 North Brazil Current rings

The extension of the NBC retroflection varies depending on the state of eddy formation. It moves northwestward up until 9°N as an eddy is forming, and then retracts to the southeastern part of the region. During our period of interest, the retroflection shed anticyclone A1 at the beginning of February. It is difficult to estimate the date of shedding as the area is highly dynamic and detecting the first closed contour of ADT is complicated and may be inaccurate. It is however interesting that the two ships sampled the retroflection when it was expanding to generate A1, and this northwestward expansion is well observed on several physical and biogeochemical parameters (Figure 7a). RV Merian crossed the retroflection on January, 27<sup>th</sup> and stayed in the area until February, 2<sup>nd</sup> (Figure 6). Chla present on the shelf is advected by the strong currents on the periphery of the retroflection and delineates well its south-western side (Figure 7a). The NBC waters stand out on the surface TS diagram, as they are the most saline waters observed in the region (Figure 7b). They are also high in fCO<sub>2</sub> which reflects their equatorial origin. Their SST is relatively warm, varying from 27.8 °C at the crossing of the first retroflection front, to 27.2 °C. The region is rather homogeneous, with an almost constant SSS of 36.3 and ΔfCO<sub>2</sub> along the multiple crossings, as observed on the Merian and Atalante transects (Figure 6). On average along those transects, the NBC fCO<sub>2</sub> is higher than fCO<sub>2atm</sub> by 20 μatm.

Anticyclone A1 is further crossed by the RV Atalante on February 6<sup>th</sup>, just a few days after its separation from the retroflection (Figure 7c). The surface signal is almost lost, both in SST and in fCO<sub>2</sub> (Figure 7c-d). It is mainly composed of modified NBC water, which properties are close to the NBC water (high SSS, high SST, low Chla) but not as pronounced. This water mass covers a larger area, which mainly encompasses the Eddy Boulevard region. It is defined here as  $35.6 < \text{SSS} < 35.6$ ,  $27.16 < \text{SST} < 27.6$  °C and  $0.11 \text{ mg m}^{-3} < \text{Chla} < 0.25 \text{ mg m}^{-3}$  (Table 1). While the high Chla water delimits well the retroflection area, it partly covers the eddy A1.

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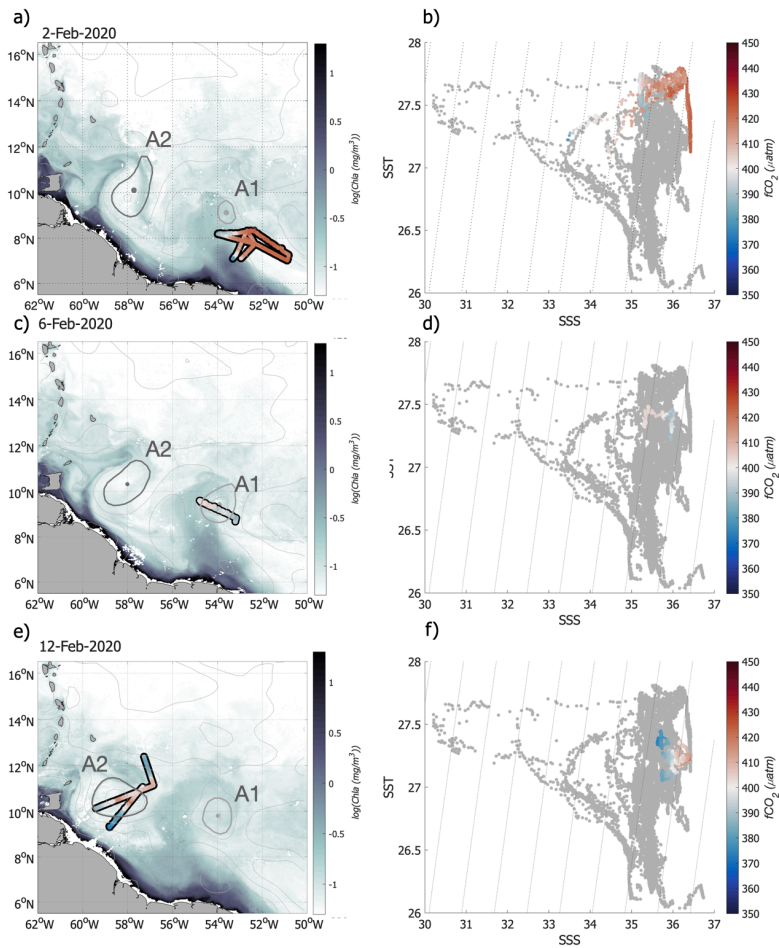


Figure 5: a) RVs Atalante and Merian ship track in the NBC retroflection (Merian: Jan. 27<sup>th</sup> to Feb. 2<sup>nd</sup>, Atalante: Feb 2<sup>nd</sup>), c) in NBC ring A1 (Atalante: Feb 6<sup>th</sup>) and e) in NBC ring A2 (Atalante: Feb 12<sup>th</sup>-13<sup>th</sup>, Merian: Feb 13<sup>th</sup>-14<sup>th</sup>), colour-coded with fCO<sub>2</sub>. The background represents the Chl a on Feb 2<sup>nd</sup> (a), Feb 6<sup>th</sup> (c) and Feb 12<sup>th</sup> (e), and the contours of NBC rings A1 and A2 are indicated. b),d),f) Corresponding T-S diagrams, colour-coded with fCO<sub>2</sub>.

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835 NBC ring A2 presents a different situation. Detached from the retroflection in early December 2019 (as defined from altimetry detection), it travelled north-westward while retaining an intense coherent core. Coastal waters identified by their high Chla content were less present and mostly entrained at the north-westward edge of the eddy. After two months, A2 almost reached Trinidad and Tobago and was located around 11°N/58°W when it was sampled by the two ships (Figure 7c). The SST signal is eroded, and most of the eddy is mostly made of modified NBC water with relatively low fCO<sub>2</sub>. However high SSS (36.5) and fCO<sub>2</sub> (415  $\mu$ atm) are still visible near the eddy ~~centre~~ on the two crossings made by RV Atalante on the 12<sup>th</sup> and 13<sup>th</sup> of February. This is confirmed by the two sections of the Merian that crossed the altimetric eddy ~~centre~~ and measured SSS 0.5 higher in the 50 km radius around the ~~centre~~ and above 36. The NBC water mass is therefore found close to the ~~centre~~ of eddy A2, as well as its associated high fCO<sub>2</sub>.

845 From the collected observations, it appears that the surface signature of NBC rings is relatively variable and complex. It is well marked in their formation area in the NBC retroflection, where waters brought north by the NBC are warmer, saltier, and higher in fCO<sub>2</sub> than the water of the northwestern tropical Atlantic. As the eddies travel northwestward, further away from the retroflection, they may be subject to various processes that modify the surface signal. Unfortunately, the data collected is not sufficient to shed light on which processes ~~are~~ involved in this situation. South of Barbados, away from the retroflection, the modified NBC is therefore the most common water mass. Nevertheless, the NBC water is still sometimes observed months after the separation from the retroflection, in the eddy ~~centre~~.

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### 3.4 Freshwater plume

The NBC rings form and evolve in an area highly influenced by the Amazon River plume. Even if February is a period of low Amazon River outflow (Dai & Trenberth, 2002), freshwater events are relatively common. In February 2020, a freshwater plume detached from the Guiana plateau and spread out into the northwestern tropical Atlantic. The off-shelf plume was steered northward by the retroflection and NBC ring A1 up to 12°N and then extended westward toward the Caribbean Sea. Waters carried by the plume strongly contrast with the saline waters of the retroflection. They include water from the Amazon and present low SSS (SSS < 34.5), low fCO<sub>2</sub> (fCO<sub>2</sub> < 380  $\mu$ atm) and high Chla (Table 1). The plume was crossed three times, twice on February 2<sup>nd</sup> and once on February 5<sup>th</sup> (Figure 6). Freshwater from the Amazon arrived on the plateau on the 1-2<sup>nd</sup> of February and was then entrained northwestward by Ekman transport and geostrophic currents (Reverdin et al., 2021). On the 2<sup>nd</sup> of February RVs Atalante and Merian left the retroflection area to cross the adjacent nascent plume. SSS rapidly decreased, reaching 33 which is associated to a strong decrease of fCO<sub>2</sub> (Figure 8). From the 2<sup>nd</sup> to the 5<sup>th</sup> of February, the plume formed and on February 5<sup>th</sup> the plume is approximately 100 km wide, with lowest salinities around 30. Based on satellite SSS data of the following days, the plume appears to have reached even lower salinities and then spread out over the north-western tropical

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875 Atlantic. It first spread northward, steered by A1 and then northwestward, ~~channelled~~ between A1 and A2 reaching all the way  
up to 12°N and extending over more than 100 000 km<sup>2</sup> (Reverdin et al., 2021). The plume can be followed by satellite SSS  
and Chla maps (Figures 7a, c, e). Indeed, the low SSS is also accompanied by high Chla as water from the Amazon are  
considered highly productive. The north-western tropical Atlantic is in general nutrient-limited but the nutrients brought by  
the Amazon can support the occurrence of a bloom. The plume is also characterized by high silicate (between 4 and 10  $\mu\text{mol}$   
880  $\text{kg}^{-1}$  in the plume, almost 0 elsewhere), while nitrate and phosphate are rapidly consumed. Traces of inorganic phosphorus  
were observed in the plume, while nitrates were absent from surface waters (Supplementary Figure S2). Low salinity combined  
with high biological productivity led to low  $f\text{CO}_2$  and a strong carbon drawdown in the plume, as the  $\Delta f\text{CO}_2$  reached  $-73 \mu\text{atm}$   
on February 5<sup>th</sup> (Figure 6).

In an area highly influenced by the NBC waters, through rings or the retroreflection, the plume stands out and ~~modifies~~ the  
885 biogeochemical dynamics of the region.

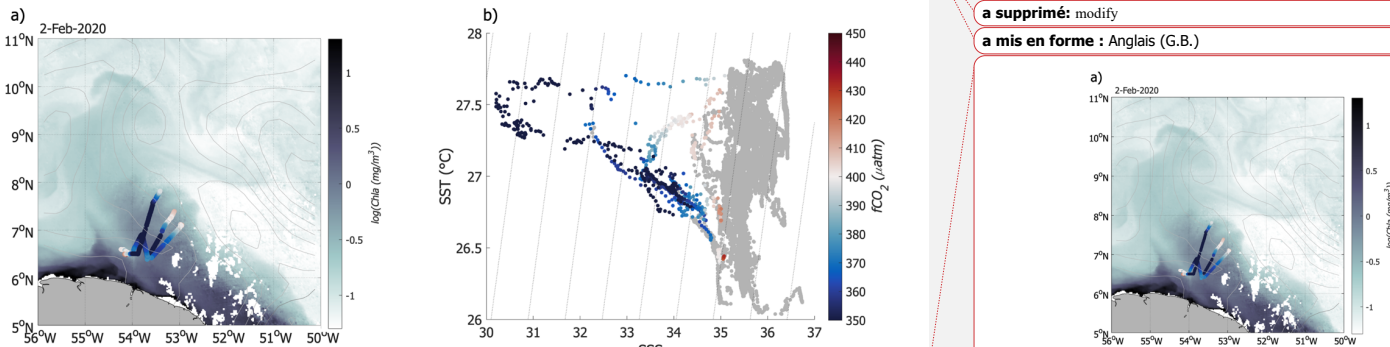


Figure 6: a) RVs Atalante and Merian ship track in the freshwater plume (Atalante: Feb 2<sup>nd</sup>, Feb 5<sup>th</sup>, Merian: Feb 2<sup>nd</sup>), colour-coded with  $f\text{CO}_2$ . The background represents the Chla on Feb 2<sup>nd</sup>. b) Corresponding T-S diagram colour-coded with  $f\text{CO}_2$ .

### 890 3.5 Shelf water and filaments

The freshwater plume is not the only water stirred by the NBC rings travelling from the NBC retroflection towards the Lesser  
Antilles. The shelf water is very different from the plume water, and was only sampled sparsely on the way in and out of the  
plume (Figures 4,5). On the Guiana plateau water is very rich in Chla and detrital material, rather saline (SSS  $\sim$  35.5) and  
relatively cold (SST  $\sim$  26.5°C) (Figure 5, Table 1). Since the water sampled on the edge of the plume was cold due to a local

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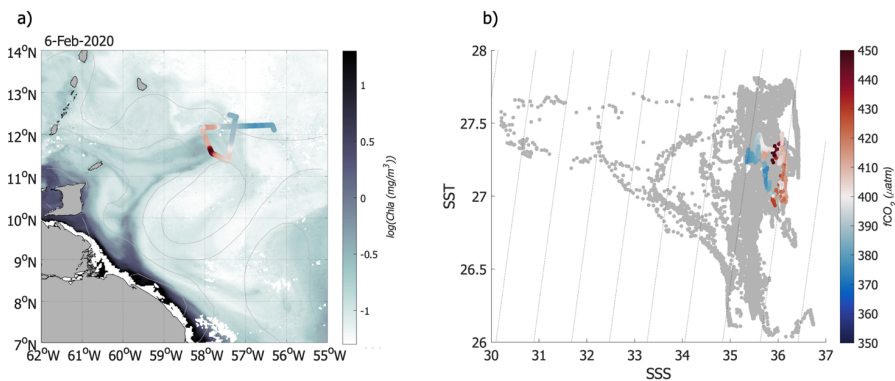
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905 upwelling event (and/or vertical mixing event) detailed in the supplementary materials (Figure S3), temperature is not homogenous on the shelf.

Further north, a filament is stirred on the western side of NBC ring A2 (Figure 9a). It is a small-scale structure, approximately 10 km wide, easily identifiable due to its high Chla. The filament is continuously stirred by A2, and so is already visible on Chla maps of February 2<sup>nd</sup> (Figure 7). It followed A2's westward translation and was crossed on February 6<sup>th</sup> and 17<sup>th</sup> by RV Merian, and on February 14<sup>th</sup> by RV Atalante (Figure 6). It has a SSS close to 36, and an SST between 27°C and 27.5°C, thus it is slightly colder and more saline than its surrounding waters (Figure 9b, Table 1). It stands out by its high Chla content (Chla > 0.25 mg m<sup>-3</sup>), even if this is lower than close to the coast or in the freshwater plume. The strongest signal is observed on the ocean carbon parameters. In contrast to the freshwater plume, this filament presents very high fCO<sub>2</sub> (> 430 μatm), highlighting different origins. It stands out from the ship track time series by also having a larger positive ΔfCO<sub>2</sub> (50 μatm).

915 Whereas the freshwater plume observed more southeastward carries water recently arrived on the plateau from the Amazon, the northwestward filament contains shelf waters.



920 **Figure 7:** a) RV Merian ship track in the shelf water filament (Feb 6<sup>th</sup>), colour-coded with fCO<sub>2</sub>. The background represents the Chla on Feb 6<sup>th</sup>. b) Corresponding T-S diagram colour-coded with fCO<sub>2</sub>.

### 3.6 Air-sea CO<sub>2</sub> flux

In order to better characterize the impact of each structure on the regional flux, we computed air-sea CO<sub>2</sub> flux maps from satellite data, at a resolution of 2.5 km (Figure 10), averaged over the period of the cruise (February 2<sup>nd</sup> to February 19<sup>th</sup>). The along-track flux represented on Figure 10a and the reconstructed regional field (Figure 10b) show the importance of the small-scale dynamical structures, and highlights two strong regimes that are found on the reconstructed map. The air-sea CO<sub>2</sub> flux

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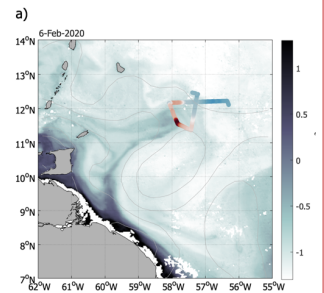
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in the northeastern part of the domain, characterized by the NASW, is mainly dominated by temperature effects while further south the presence of NBC rings, and their interactions with coastal waters, create a strong dependence of the CO<sub>2</sub> flux on SSS and on the biological and biogeochemical processes highlighted by the Chl<sub>a</sub>.

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We evaluate the integrated air-sea CO<sub>2</sub> flux over the region. In February, waters are the coldest, and the region is a strong CO<sub>2</sub> sink of -1.7 TgC month<sup>-1</sup> (Figure 11). Three biogeochemical domains mainly contribute to the air-sea CO<sub>2</sub> flux, the NASW, the freshwater plume and the NBC retroflection. The impact on the flux of the small-scale coastal filament is evident along the ship tracks (Figure 10b). However, its contribution to the total flux is weak as the signal is smoothed when averaging over February 2<sup>nd</sup> to 19<sup>th</sup>, as the filament moves following the A2 ring northwestward translation. Each of the main three regions is identified based on its averaged SST, SSS and Chl<sub>a</sub> properties in February and the region-specific flux is determined (Figure 11).

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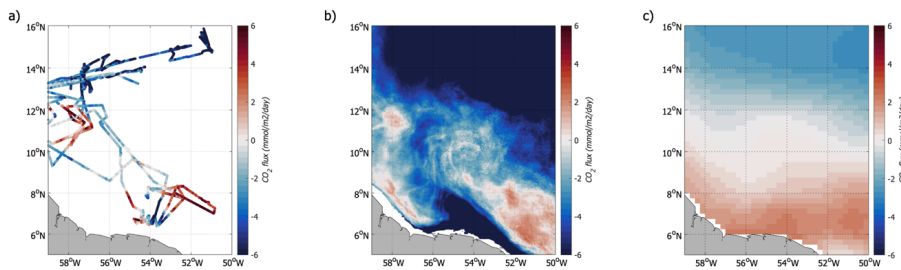


Figure 8: a) Air-sea CO<sub>2</sub> flux measured in Jan-Feb 2020 during the EUREC<sup>4</sup>A-OA/ATOMIC cruise. b) Air-sea CO<sub>2</sub> flux reconstructed over February 2020. c) February climatology of the air-sea CO<sub>2</sub> flux over 1998-2015 (Landschützer et al., 2020).

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NASW contributes to about 60% of the total sink due to their relatively cold temperature and to strong winds that enhances the air-sea exchanges. These waters extend from Barbados northward and eastward, cover more than 1/3 of the domain and show a weak variability over February 2<sup>nd</sup> to 19<sup>th</sup>.

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The NBC retroflection is a source of CO<sub>2</sub> to the atmosphere. In February, the strongest signal is observed in the southeastern part of the domain up to 8°N/53°W. The retroflection nevertheless impacts the region as far as 10°N/54°W as it is spatially variable, reaching up to 10°N when shedding an eddy. The two NBC rings present a small positive February air-sea CO<sub>2</sub> flux average. Eddy A1 is almost stationary from its formation date (around February 6<sup>th</sup>) until February 20<sup>th</sup> and its neutral to

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slightly positive CO<sub>2</sub> flux is ~~centred~~ around 10°N/54.5°W (Figure 10). Eddy A2 translates rapidly westward at the beginning of February, and then northward from the 15<sup>th</sup> of February. Its signal is therefore not as visible as on the ship tracks as it is averaged over ~~19 days~~. The retroflection is the main region with a positive air-sea CO<sub>2</sub> flux, even if the region is too small to have a big global impact, understanding small-scale features may be significant for the total flux. The NBC rings carry part of the signal, which is heavily modified as they travel northwestward. As a result, on average ~~in early to late~~ February only the retroflection maintains a positive flux, while a large part of the domain dominated by modified NBC waters (non-influenced by the plume) behaves as a small sink.

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The freshwater plume with Amazon water is nascent when crossed by the ships (Figure 10b), but is already the strongest signal of the time-series. As the plume develops, it is entrained by NBC ring A1, then A2 and spreads out into the open ocean, as observed on SSS and Chla maps. The plume generates a strong CO<sub>2</sub> sink that is amplified by strong winds, and reaches up to 12°N (Figure 10). The freshwater plume covers only 10% of the total area, but contributes to almost 20 % of the sink. In winter, this region is either not characterized in previous studies, or considered as dominated by high fCO<sub>2</sub> waters brought by the NBC on the climatology. We observe here that the ~~trapping of CO<sub>2</sub>-rich water by NBC rings effect~~ is relatively weak in winter, and the main signal is associated to the filaments they stir.

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The north-western tropical Atlantic therefore behaves as a sink of CO<sub>2</sub> in ~~early to mid~~-February, driven by the cold north Atlantic subtropical waters and the Amazon freshwater plume stirred by NBC rings.

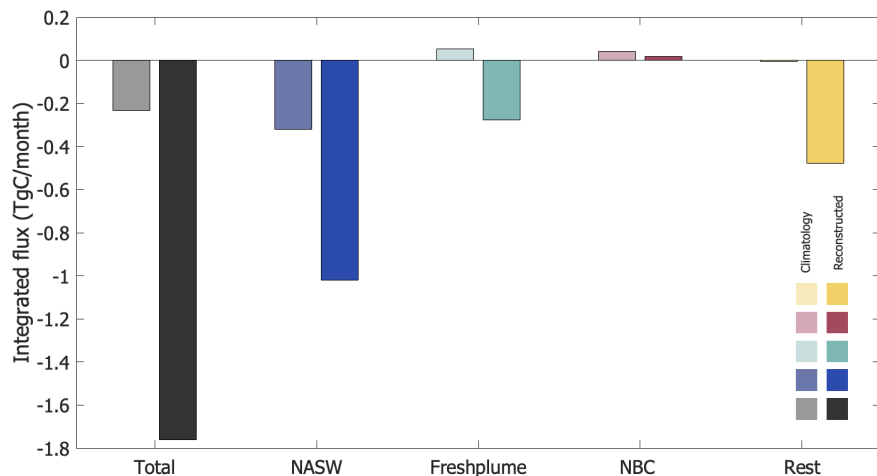


Figure 9: Integrated flux for the [5°-16°N, 59-50°W] domain, and for 3 water masses. For each bar duet, the one on the left in faded colours represents the integrated flux from Landschützer et al., (2020) February climatology, while the one on the right is computed from the reconstructed flux. Same colour code as in Figure 5.

## 4 Discussion

### 4.1 Integrated air-sea CO<sub>2</sub> flux

The northwestern tropical Atlantic presents a strong seasonal variability of air-sea CO<sub>2</sub> fluxes (Landschützer et al., 2016). In February, waters are the coldest, and we estimate the 5-16N, 59-50W domain to be a strong CO<sub>2</sub> sink of -1.7 TgC month<sup>-1</sup> (Figure 10). This region, located at tropical latitudes but combining characteristics of subtropical waters and river outflow, is difficult to represent in large-scale climatologies. Indeed, the sink for the month of February is smaller by a factor 10 in Landschützer et al. (2020), and is also considerably smaller in Takahashi et al., (2009), but the low spatial resolution of this last product doesn't allow for a good quantitative comparison. This region has been rarely observed, and the interannual variability described in Landschützer et al., (2020)'s climatology is therefore rather uncertain. The compensating effect of different years cannot explain entirely the difference of signal observed in February 2020 with respect to the two climatologies.

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- In an effort to understand how biogeochemistry is forced by physical processes in the ocean and atmosphere, we used surface Chl<sub>a</sub> to complement SST and SSS data in defining surface water masses. Partitioning the world ocean into biogeochemical provinces based on physical, geographical and biological criteria has long been proposed to study and better understand biological processes (Fay & McKinley, 2014; Longhurst et al., 1995... [73])
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085 Three water masses mainly contribute to the air-sea CO<sub>2</sub> flux, the NASW, the fresh plume and the NBC retroflection. The NASW contributes to about 60% of the total sink and is not well captured in climatologies, with noticeable differences of more than 20 μatm between the measured ΔfCO<sub>2</sub> in 2020 and the one computed from Landschützer et al., (2020) and Takahashi et al., (2009) (the closest grid point is considered for this comparison). The retroflection is the main region with a positive air-sea CO<sub>2</sub> flux. Its influence is observed up to 10°N-55°W, but its area is small so its impact on the regional flux is weak. The positive flux of the retroflection is slightly overestimated in the climatologies, but it could also be due to the difficulty to detect the retroflection at the beginning of February. The main difference is that the NBC waters rich in CO<sub>2</sub> are localized in the retroflection area, and are heavily modified when spreading into the Eddy Boulevard.

090 The freshwater plume is a feature previously not well described for this region in winter and we found a contribution of almost 20 % to the sink. The impact of the Amazon River has been overlooked so far in winter, but it accounts for a large part of the salinity and biogeochemical variability. Freshwater from the Amazon is not just located on the shelf, but it can spread northward advected by the strong current variability associated to the NBC rings (Reverdin et al., 2021). These rings are the largest, faster rotating and are the most energetic during boreal winter compared to other seasons (Aroucha et al., 2020). Combined with a seasonal increase in the Amazon's outflow, it induces a large variability in SSS, Chl<sub>a</sub> and fCO<sub>2</sub>. The occurrence of freshwater export from the shelf to the open ocean has a strong influence on the salinity and therefore on the mixed layer depth and air-sea heat exchanges (Reverdin et al., 2021). It also strongly impacts the biogeochemistry of the region as the low fCO<sub>2</sub> is due both to the low salinity of the plume waters and to the biological activity. The plume stirred into the open ocean by the NBC rings brings nutrients in a region strongly nutrient-limited, and generates a local winter bloom. This in turns plays an important role on the air-sea CO<sub>2</sub> flux, and is a crucial feature of the southern part of the northwestern tropical Atlantic.

#### 4.2 Extension to other years and interannual variability

095 Few tropical Atlantic measurements of biogeochemical tracers are available, in particular in the northwestern tropical Atlantic. The EUREC<sup>4</sup>A-OA/ATOMIC campaign provides the first in-situ comprehensive measurements of fCO<sub>2</sub> in this region for the boreal winter season. The reconstruction of fCO<sub>2</sub> maps likely provides a good understanding of the spatial evolution of fCO<sub>2</sub> and air-sea CO<sub>2</sub> fluxes, and is fitted for the months of January-February 2020. Although the processes described here are specific to winter and thus cannot be extended to other seasons, they will be useful to understand the winter variability of other years.

110 Only a few cruises cross the region according to the SOCAT database between 2010 and 2019 (period with satellite SSS data) and investigating inter-annual variability is not possible. However, we can test the relation developed for 2020 for other years by using selected cruises from the SOCAT database. We thus first use the relationship to reconstruct fCO<sub>2</sub> along the ship tracks (using in-situ SSS and SST and colocalized Chl<sub>a</sub>) and then over the whole region based on satellite products (OSTIA SST, Globcolour Chl<sub>a</sub>, SSS+CCI, detailed in the appendix A). A comparison between the measured and reconstructed fCO<sub>2</sub> for the water masses sampled by the SOCAT cruises (NASW, fresh plume, NBC retroflection, modified NBC) is presented in the

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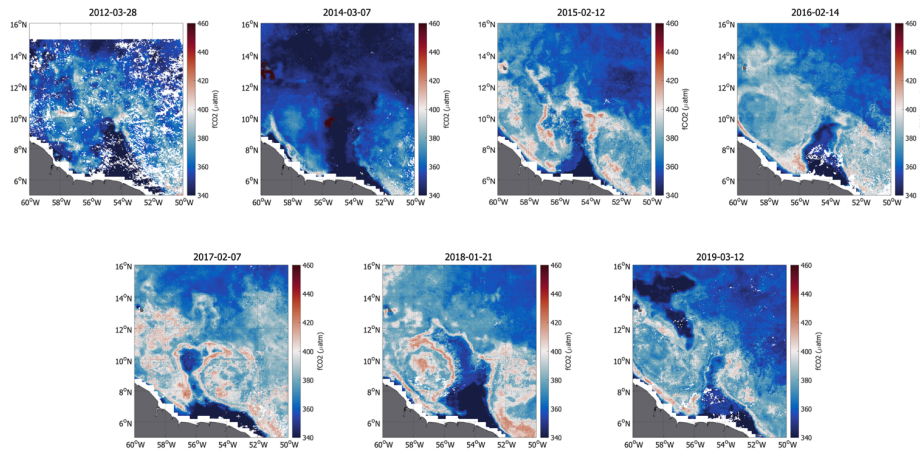
appendix (Table A1). Good agreement is found between the  $f\text{CO}_2$  from the SOCAT database and the one reconstructed from the in-situ temperature and salinity, and colocalized  $\text{Chl}a$  for the four water masses (averaged difference of  $5.5 \mu\text{atm}$ ). When comparing reconstructed  $f\text{CO}_2$  maps with  $f\text{CO}_2$  on ship tracks (Figure A1), the agreement between  $f\text{CO}_2$  in various water masses is very clear, even though the spatial structures are sometimes a bit misplaced. This is attributable to the slightly coarser resolution of satellite products not designed specifically for each campaign, to the high spatio-temporal variability of  $f\text{CO}_2$  and to missing  $\text{Chl}a$  and SST observations in cloudy areas. February 2020 was mainly cloud free, so that we were able to use high resolution daily SST and  $\text{Chl}a$ . The SSS product used in 2020 is also a daily product. However, for the other years, the satellite  $\text{Chl}a$  (if clouds) and SSS products have a weekly temporal resolution, which smear the fast-moving structures. The gradients between water masses are therefore not always well represented, but we find a good agreement between the  $f\text{CO}_2$  of each structure, which is encouraging for future studies on interannual variability in winter.

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**Figure 10:** Snapshot of reconstructed  $f\text{CO}_2$  for all occurrences of fresh plumes extending at least to  $10^\circ\text{N}$  and east of  $56^\circ\text{W}$  in January-March 2010-2019 (2010, 2011 and 2013 do not present this type of event).

By identifying the main processes responsible for the variability of the air-sea  $\text{CO}_2$  flux in 2020, we can better understand the interannual variability of the region. Indeed, each of the main water mass has its own interannual variability that shapes the  $\text{CO}_2$  variability. The northern part of the domain is dominated by the variation of temperature, and therefore its interannual variability is mainly linked to the one of SST. From 32 years monthly mean SST data, the SST standard deviation in the area

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is relatively weak, and doesn't exceed 0.5°C. The northern sink of CO<sub>2</sub> is therefore rather similar from year to year, coherent with the low standard deviation of the air-sea CO<sub>2</sub> flux computed from Landschützer et al., (2020). Some variability is still observed on the snapshot of the reconstructed fCO<sub>2</sub> (Figure 12), but to a much smaller extent than south of Barbados. For example, the strong sink observed in March 2014 is caused by cold SST anomalies over the whole domain. Some small-scale variability in the northern part of the domain is sometimes correlated to SSS anomalies, as in 2019.

The freshwater plume sampled during EUREC<sup>4</sup>A-OA is a common feature in February. During the 2010-2019 period, events of freshwater reaching the open ocean were observed each year, and freshwater plumes similar to the one described in this paper were observed during 7 out of 10 years from satellite salinity data (Reverdin et al., 2021). Two of the main mechanisms driving the occurrence of the plume are the winds near the Amazon estuary that can induce along shelf transport to the Guyana plateau and the presence of NBC rings. Most of the plume events similar to the one in this study suggest the presence of an anticyclone to its east. This region is commonly crossed by several NBC rings during winter (Jochumsen et al., 2010; Johns et al., 2003; Mélice and Arnault, 2017) but it also is subject to a strong year to year variability that has linkages with the variability of the Amazon River outflow (Aroucha et al., 2020). Therefore, identifying and understanding the processes happening in 2020 should contribute to better assess the interannual variability of fCO<sub>2</sub> as well as air-sea CO<sub>2</sub> fluxes in the northwestern tropical Atlantic during winter. Using a combination of SSS, SST and Chl<sub>a</sub> brings information on the biogeochemistry of the area in winter and represent well the mesoscale structure.

## 5. Conclusion

The EUREC<sup>4</sup>A-OA/ATOMIC campaign provides for the first time synoptic measurements related to the air-sea fluxes of CO<sub>2</sub> in the northwestern tropical Atlantic in winter. Six main surface water masses are identified, one of them north of Barbados (North Atlantic Subtropical Water), and the other five (the NBC retroflection, modified NBC waters, the freshwater plume, the shelf water and the shelf filament) south of Barbados. The investigation highlights the two different regimes of the region. In the northern part, the variability of the CO<sub>2</sub> flux is low and the area is covered by relatively cold, saline and low-chlorophyll NASW. The southern part is highly variable, due to the presence of large mesoscale anticyclonic eddies. In January and February 2020, two NBC rings influence the physical and biogeochemical properties of the region. The NBC retroflection is characterized by waters with equatorial origins that are relatively warm, saline and high in fCO<sub>2</sub>. As the rings separate from the retroflection, they interact with the surrounding waters, and the initial signal in fCO<sub>2</sub> is dampened. The main impact of the rings is therefore not necessarily on the surface water they transport in their core (eddy trapping), but rather on the filaments they stir off the coast (eddy stirring). A fresh plume from the Amazon River is transported by the coastal current up to the French Guiana shelf in the beginning of February. The NBC rings entrain the plume of freshwater up to 12°N. This plume is fresh, rich in Chl<sub>a</sub> and low in fCO<sub>2</sub> and strongly contrasts with the surrounding waters and spreads over ~100 000 km<sup>2</sup>. On the shelf not influenced by the plume, water is relatively saline, high in fCO<sub>2</sub> and Chl<sub>a</sub>, probably due to high concentration of

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a supprimé: As previously stated, most of the variability occurs south of Barbados.

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a supprimé: This region is commonly crossed by several NBC rings during winter (Jochumsen et al., 2010; Johns et al., 2003; Mélice & Arnault, 2017)

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detrital material. As ring A2 propagates westward, it continuously stirs a thin (10 km wide) filament of high fCO<sub>2</sub> shelf water up to 12°N.

185 Based on the ship observations we identify distinct regimes in fCO<sub>2</sub> linked to certain combinations of SST, SSS, and Chl<sub>a</sub> properties. We use this information to construct high-resolution maps of fCO<sub>2</sub> and air-sea CO<sub>2</sub> flux using satellite maps of SSS, SST and Chl<sub>a</sub>. On average over early to mid-February, the region acts as a strong sink of CO<sub>2</sub> (-1.7 TgC<sub>2</sub>month<sup>-1</sup>), the sink being ten times smaller in air-sea CO<sub>2</sub> flux climatologies. The NASW is responsible for most of the flux (60%) due to low temperature associated to winter cooling and strong winds. South of Barbados, the region acts also as a sink of CO<sub>2</sub>. The influence of equatorial water is localized to the retroreflection region that acts as a small source of CO<sub>2</sub>. The main feature in this part of the domain is the fresh plume that contributes almost 20% of the total sink.

190 The processes described here highlight the high variability of air-sea CO<sub>2</sub> fluxes in winter, that are quite different from the ones in summer. These features are relatively common in winter and can be used to better understand the interannual variability of air-sea CO<sub>2</sub> fluxes. The northern part of the domain is driven by the variability in SST, while the southern one is a combination of the interannual variability of temperature, salinity and chlorophyll. It is therefore linked to the year-to-year variability of the NBC rings and the Amazon outflow.

195 This study is limited by the paucity of data in the region and for this time period. More fCO<sub>2</sub> data closer to the coast would help to better quantify the influence of shelf water on the flux. The signature of the NBC rings has been described for only two rings that had different signatures. In order to reach more robust conclusions on the transport of surface NBC water by the rings, more eddies should be observed. The variability of fCO<sub>2</sub> occurs at large and small scale. Salinity is one of the most valuable predictors of fCO<sub>2</sub> south of 10°N, but the satellite salinity resolution is much lower than those of temperature and chlorophyll. To have a more accurate prediction of the fCO<sub>2</sub>, a high-resolution SSS product would also be very useful.

## Appendix A

205 Due their long time series, the following SST, Chl<sub>a</sub> and SSS products are used to reconstruct fCO<sub>2</sub> maps in winter in the northwestern tropical Atlantic for other years than 2020. Results are shown on Figure A1, and Figure 12. They are different than the satellite products used in the main study, that were only available on a short period.

The Operational Sea Surface Temperature and Sea Ice Analysis (OSTIA) SST product, distributed by the CMEMS is used here. Daily maps of SST are produced at a resolution of 1/20°, available from 1981 to present. OSTIA SST uses most SST data available for a day, from both infrared and microwave inferred SST.

210 Surface Chl<sub>a</sub> from GlobColour dataset derived from ocean color at a 1/24° resolution is used. It is a merged product from multiple satellite missions' observations (SeaWiFS, MERIS, MODIS, VIIRS NPP, OLCI-A, VIIRS JPSS-1 and OLCI-B). GlobColour data is developed and validated by ACRI-st and distributed by the CMEMS.

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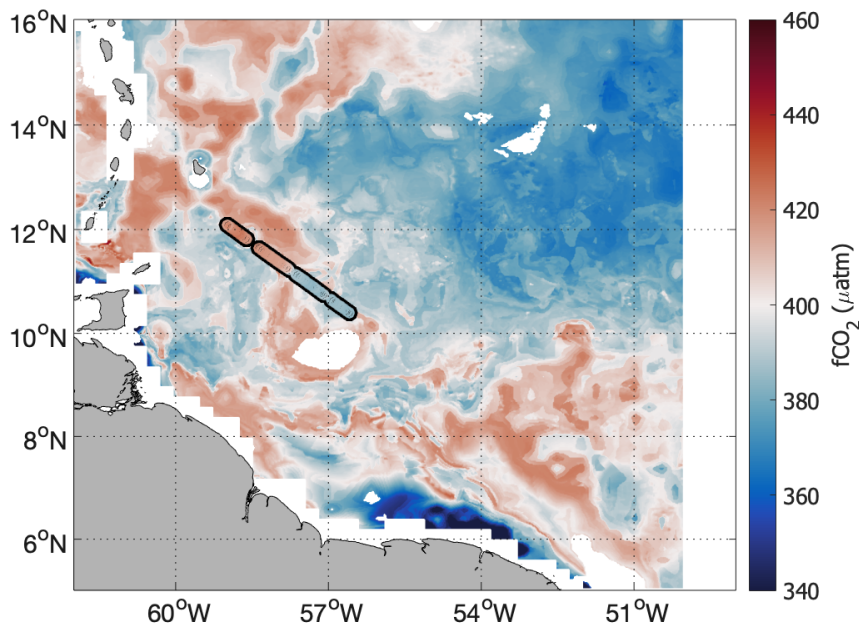
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We also use SMOS and SMAP combined weekly SSS generated by the Climate Change Initiative Sea Surface Salinity (CCI + SSS) project (Boutin et al., 2021, <http://dx.doi.org/10.5285/5920a2e77e3c45339477acd31ce62e3c>). It provides weekly level-3 SSS data from 2010 to 2019 at a spatial resolution of 50 km, a sampled daily on a 25 km x 25 km grid, by combining data from the SMOS, Aquarius, and SMAP missions.

**a supprimé:** We also use SMOS and SMAP combined weekly SSS generated by the Climate Change Initiative Sea Surface Salinity (CCI + SSS) project (<https://doi.org/10.5285/4ce685bff631459fb2a30faa699f3fc5>). It provides weekly level-3 SSS data from 2010 to 2019 at a spatial resolution of 50 km, a sampled on a 25 km x 25 km grid, and 1 day, by combining data from the SMOS, Aquarius, and SMAP missions.¶  
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▲ Figure A1. fCO<sub>2</sub> reconstructed from OSTIA SST, CCI+SSS and Globcolour Chla for the 23/12/2015 superimposed with the fCO<sub>2</sub> from cruise 642B20151209.

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	Fresh plume	NBC waters	Modified NBC	NASW
SOCAT fCO <sub>2</sub>	316.2	413.4	385.8	349
fCO <sub>2</sub> reconstructed from SOCAT SST & SSS	310.7	410.7	392.9	358.7
Transect date	2016/01/05	2016/01/08	2015/12/23	2013/02/10
Ship Name	Colibri (France)	Colibri (France)	MSC Marianna (Panama)	Benguela Stream (Netherlands)
Expocode	35MJ20151229	35MJ20160107	642B20151209	33RO20130108

Table A1. Comparison for the 4 main water masses between the fCO<sub>2</sub> from SOCAT transect and the fCO<sub>2</sub> reconstructed from in-situ SSS and SST and colocalized Chla.

#### Code Availability

Code used in this study can be made available upon reasonable request to the corresponding author.

#### Data Availability

We benefited from numerous data sets made freely available and listed here: the ADT produced by Ssalto/Duacs distributed by CMEMS (<https://resources.marine.copernicus.eu>), the Chla and SST maps produced by CLS (<https://datastore.cls.fr/catalogues/chlorophyll-high-resolution-daily> and <https://datastore.cls.fr/catalogues/sea-surface-temperature-infra-red-high-resolution-daily>), the SMOS L2Q field produced by CATDS (CATDS, 2019) (<https://10.12770/12dba510-cd71-4d4f-9fe1-9cc027d128b0>), the SMAP maps produced by Remote Sensing System (RSS v4 40 km), the CCI+SSS maps produced in the frame of ESA CCI+SSS project (<http://dx.doi.org/10.5285/5920a2c77e3c45339477acd31ce62c3c>), the OSTIA SST and Copernicus -GlobColour Chla distributed by the CMEMS (SST\_GLO\_SST\_L4\_REP\_OBSERVATIONS\_010\_011 and OCEANCOLOUR\_GLO\_CHL\_L4\_REP\_OBSERVATIONS\_009\_082).

The RV Atalante fCO<sub>2</sub> is available on the SEANOE website: doi/10.17882/83578. The RV Ron Brown and RV Merian fCO<sub>2</sub> data can be found on the SOCAT database (expocodes 33RO20200106 and 06M220200117 respectively). The Surface Ocean CO<sub>2</sub> Atlas (SOCAT) is an international effort, endorsed by the International Ocean Carbon Coordination Project (IOCCP), the

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Surface Ocean Lower Atmosphere Study (SOLAS) and the Integrated Marine Biosphere Research program, to deliver a uniformly quality-controlled surface ocean CO<sub>2</sub> database. The many researchers and funding agencies responsible for the collection of data and quality control are thanked for their contributions to SOCAT.

#### Author contribution

LO, JB, GR and NL conceptualized the project. LO carried out the measurements and data analysis. LO, JB, GR, NL and PL contributed to result interpretation. PL, MR and RW provided the crucial datasets. LO, MR, SS, JK, ML, TS and CN conducted field work. LO wrote the manuscript with input from all co-authors.

#### Competing interests

Some authors are members of the editorial board of Biogeosciences. The peer-review process was guided by an independent editor, and the authors have also no other competing interests to declare.

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