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# SCIENTIFIC REPORTS



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## Collapse of the tropical and subtropical North Atlantic CO<sub>2</sub> sink in boreal spring of 2010

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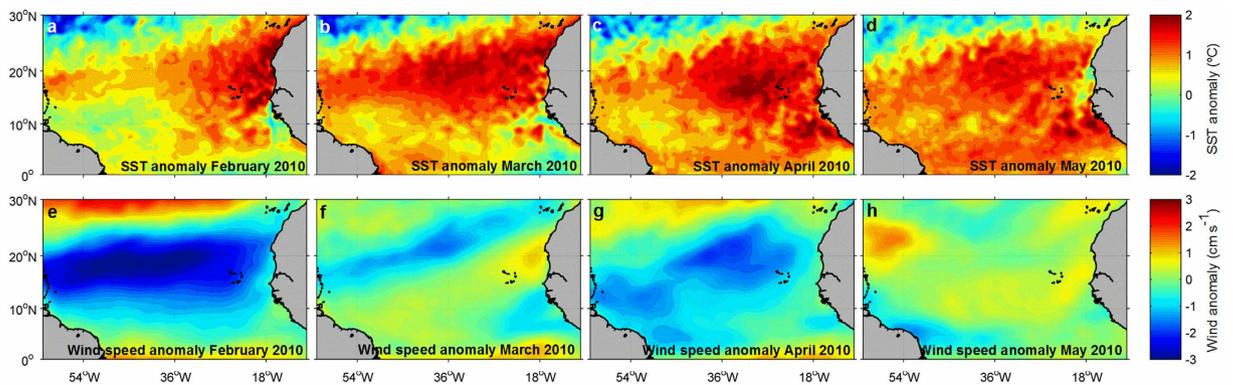
Following the 2009 Pacific El Niño, a warm event developed in the tropical and subtropical North Atlantic during boreal spring of 2010 promoted a significant increase in the CO<sub>2</sub> fugacity of surface waters. This, together with the relaxation of the prevailing wind fields, resulted in the reversal of the atmospheric CO<sub>2</sub> absorption capacity of the tropical and subtropical North Atlantic. In the region 0–30°N, 62–10°W, this climatic event led to the reversal of the climatological CO<sub>2</sub> sink of –29.3 Tg C to a source of CO<sub>2</sub> to the atmosphere of 1.6 Tg C from February to May. The highest impact of this event is verified in the region of the North Equatorial Current, where the climatological CO<sub>2</sub> uptake of –22.4 Tg for that period ceased during 2010 (1.2 Tg C). This estimate is higher than current assessments of the multidecadal variability of the sea-air CO<sub>2</sub> exchange for the entire North Atlantic (20 Tg year<sup>–1</sup>), and highlights the potential impact of the increasing occurrence of extreme climate events over the oceanic CO<sub>2</sub> sink and atmospheric CO<sub>2</sub> composition.

Anthropogenic CO<sub>2</sub> emission to the atmosphere is widely considered the main cause of current climate change. Since the industrial revolution, the oceans have absorbed about 40–50% of all the anthropogenic CO<sub>2</sub> emissions<sup>1,2</sup>, thus mitigating its effects over the Earth climate system. Nevertheless, studies have suggested that the oceanic C sink may be decreasing for the last 50 years<sup>3,4</sup>. Whether these changes are caused from anthropogenic climate change or internal climate variability is still uncertain<sup>4–6</sup>, but they could significantly impact future atmospheric CO<sub>2</sub> levels.

The North Atlantic north of 18°N is one of the oceanic regions of strongest CO<sub>2</sub> uptake (420 ± 110 Tg C y<sup>–1</sup>) representing 30% of the global oceanic CO<sub>2</sub> sink<sup>7</sup>, and an estimated interannual and multidecadal CO<sub>2</sub> uptake variability of 20 Tg C yr<sup>–17–9</sup>. The area of the North Atlantic with CO<sub>2</sub> uptake that is most sensitive to climate forcing (changes in sea surface temperature (SST) and wind speed) is the subtropical North Atlantic<sup>10</sup>. There, the sea-air CO<sub>2</sub> exchange is mainly controlled by sea surface temperature (SST) changes due to its permanent oligotrophic conditions outside upwelling areas, thus presenting the strongest seasonal variability in the sea-air CO<sub>2</sub> exchange of this ocean<sup>10</sup>. Recent warming identified in the region, partially linked to anthropogenic forcing, is already reducing its CO<sub>2</sub> uptake<sup>11</sup>.

Large-scale climate modes such as the North Atlantic Oscillation (NAO), the Atlantic Multidecadal Oscillation (AMO) and the El Niño–Southern Oscillation (ENSO) can mitigate or exacerbate anthropogenic-driven SST increase in the North Atlantic and its effects over the sea-air CO<sub>2</sub> exchange<sup>12</sup>. In 2009, a strong El Niño event occurred in the Pacific. ENSO events are known to promote positive SST anomalies in the northern tropical Atlantic through a teleconnection driven through the troposphere with a time lag of a few months<sup>13</sup>. In boreal spring of 2010, this event coincided with a strong positive AMO resulting in a strong positive SST anomaly associated with negative wind speed anomalies in the tropical Atlantic<sup>14,15</sup> (Fig. 1). Its impact in the equatorial Atlantic sea-air CO<sub>2</sub> exchange has been explored by Lefèvre *et al.*<sup>14</sup>, who showed that during this period the Intertropical Convergence Zone (ITCZ) was shifted northward compared to its climatological position, associated with a significant reduction of rainfall. The CO<sub>2</sub> undersaturation promoted by the intense rainfall associated with the ITCZ was thus significantly reduced and, consequently, CO<sub>2</sub> outgassing in this area increased. As their study mainly focused on the Western equatorial Atlantic, the impact of this climatic event in the sea-air CO<sub>2</sub> exchange in the North Atlantic is currently unknown.

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**Figure 1.** Basin-scale SST (a to d) and wind speed (e to h) anomalies during February to May 2010. SST and wind speed anomalies were calculated as the deviation of the monthly data from the climatological SST and wind speed calculated for the period 1994–2013. Monthly SST was calculated from the daily NOAA optimum interpolation (OI) SST V2 data (0.25° resolution; NOAA/OAR/ESRL PSD, Boulder, Colorado, USA; <http://www.esrl.noaa.gov/psd/>). Wind speed was obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) monthly reanalysis data set (ERA-interim, 0.25° resolution). The maps were generated with Matlab 2016a, the M\_Map 1.7 toolbox (<https://www.eoas.ubc.ca/~rich/map.html>) and using the ETOPO2 Global 2 Arc-minute Ocean Depth and Land Elevation (National Geophysical Data Center/NESDIS/NOAA/U.S. Department of Commerce, 2001. ETOPO2, Global 2 Arc-minute Ocean Depth and Land Elevation from the US National Geophysical Data Center (NGDC). Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory. <http://dx.doi.org/10.5065/D6668B75>. Accessed 1/03/2015).

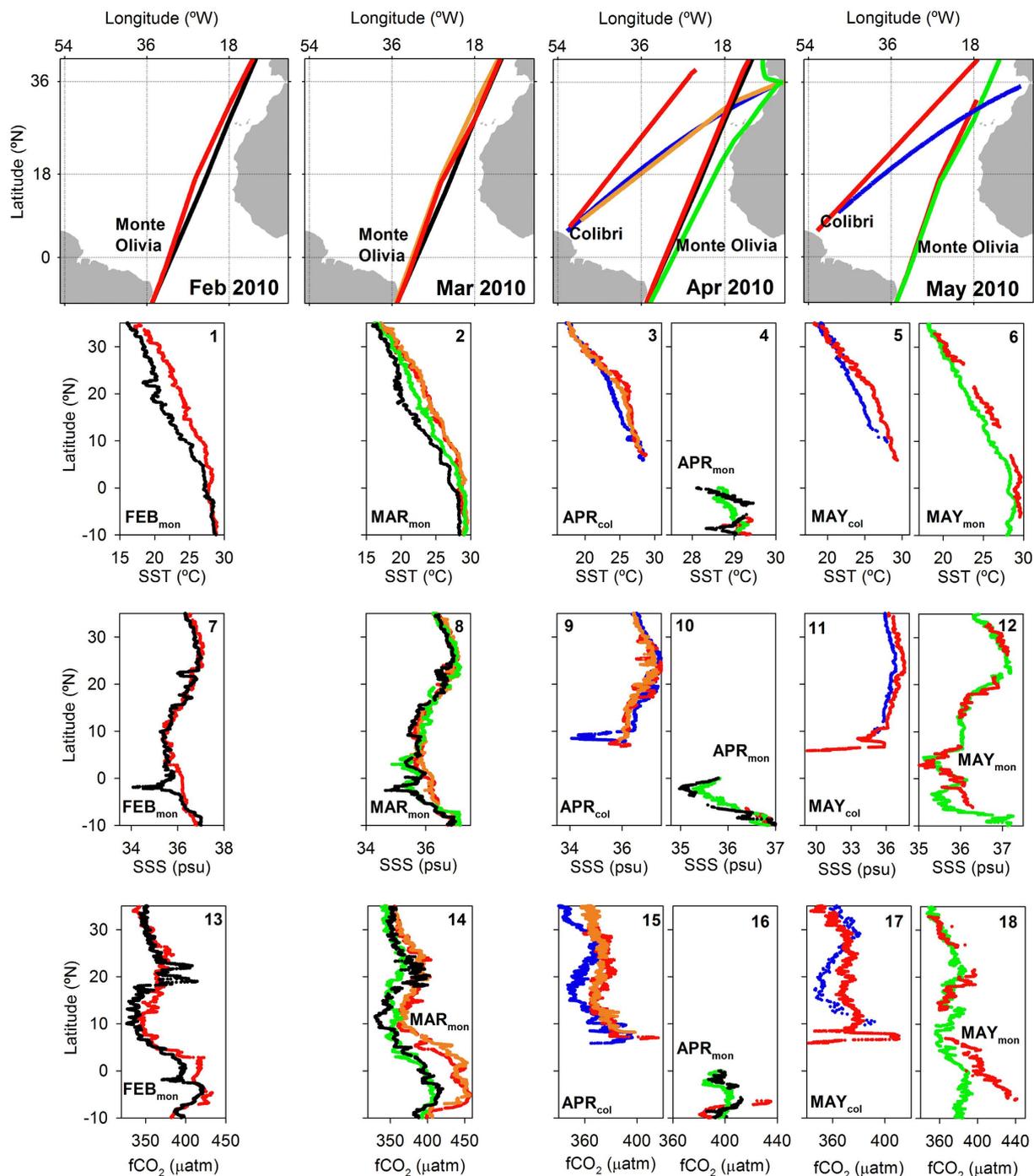
We have analyzed synoptic underway data collected onboard two Voluntary Observing Ships (VOS) that sailed across the North Atlantic during boreal spring 2010 (Monte Olivia/Rio Blanco sailing the route France-Brazil and Colibri sailing the route France-French Guyana; see Fig. 2 upper panels and Supplementary Table S1 for details on the route of the vessels). We have also used the data collected onboard these VOS during boreal spring of other years for comparison. The aim of this study is to identify the extension and quantify the impact of the SST anomaly observed in the North Atlantic climatic event of boreal spring 2010 (Fig. 1) with regard to the CO<sub>2</sub> uptake capacity of the basin.

## Results and Discussion

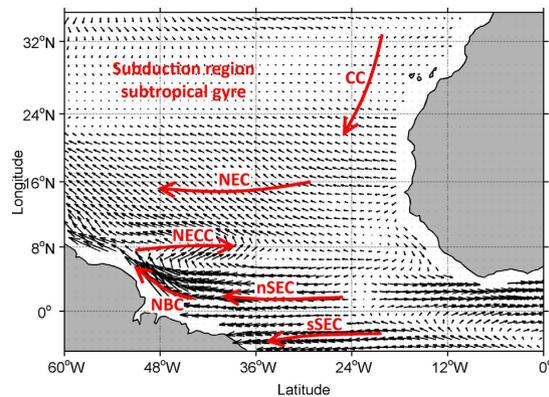
**Latitudinal distribution of sea surface temperature, salinity and the fugacity of CO<sub>2</sub>.** Underway SST measured during 2010 in the North Atlantic is significantly higher than that measured in other years along both the Monte Olivia/Rio Blanco and the Colibri tracks (differences up to 3.4°C during March; Fig. 2.2). These SST differences are registered from ~30°N to the equator along the Monte Olivia/Rio Blanco tracks and to ~10°N along the route of the Colibri. The higher underway SST measured during 2010 is accompanied by a significant increase in the measured sea surface CO<sub>2</sub> fugacity (fCO<sub>2sw</sub>). The thermodynamic effect of SST changes on fCO<sub>2sw</sub> results in ~4% fCO<sub>2sw</sub> increase for every °C due to the reduction of CO<sub>2</sub> solubility in seawater at increasing temperatures<sup>16</sup>. This thermodynamic effect of temperature on fCO<sub>2sw</sub> is well exemplified in the fCO<sub>2sw</sub> data from 2010.

Latitudinal sea surface salinity (SSS) differences among years are associated with the main freshening sources to the basin, i.e. the Amazon River plume (in the southernmost part of the Colibri tracks)<sup>17</sup> and the ITCZ (in the Southern Hemisphere along the Monte Olivia/Rio Blanco tracks)<sup>14</sup>. These freshening sources to the basin promote a lowering of fCO<sub>2sw</sub> and thus explain the fCO<sub>2sw</sub> variability in these regions (e.g. Fig. 2.14,2.17). Nevertheless, the mechanisms by which these two freshening sources affect the measured fCO<sub>2sw</sub> are different. The main process explaining the lowering in the fCO<sub>2sw</sub> in the Amazon plume compared to the surrounding oceanic waters is the C drawdown caused by the intense primary production associated with its waters<sup>17</sup>. In the case of the effect caused by the intense rainfall associated with the ITCZ, chemical dilution of total alkalinity (TALK) and dissolved inorganic C (DIC) and atmospheric DIC deposition are the main effects over the carbonate system of the oceanic surface waters. Since rainfall commonly contains zero TALK and near zero DIC, the overall effect on fCO<sub>2sw</sub> is to lower its values<sup>18</sup>.

We also observe latitudinal SSS gradients during February and March 2009 at the ~24–18°N latitudinal band along the Monte Olivia/Rio Blanco tracks, concomitant with rapid changing latitudinal SST and fCO<sub>2sw</sub> (Fig. 2). In these two cruises, measured fCO<sub>2sw</sub> was similar or even higher than that registered during 2010 despite the lower SST. This area, locus of the Canary Current (CC), presents a complex system of mesoscale features such as eddies and fronts and is locus of one of the four main Eastern Boundary Upwelling Systems<sup>19</sup>. The sharp latitudinal SST, SSS and fCO<sub>2sw</sub> gradients present in these two cruises (February and March 2009) may be associated with upwelling waters and mesoscale transport variability in the area which would explain the high fCO<sub>2sw</sub> values registered in this region.



**Figure 2. Underway measurements performed onboard the Monte Olivia/Rio Blanco/Colibri used in this study.** Upper panels show the route followed by each cruise across the North Atlantic. The latitudinal distribution of underway SST (panels 1–6), SSS (panels 7–12) and fCO<sub>2sw</sub> (panels 13–18) for the voyages used here are shown. Color code denotes the year of each cruise: blue represents 2007, black 2009, red and orange 2010, and green 2011. Subscript “mon” denotes data collected onboard the Monte Olivia/Rio Blanco, while subscript “col” refers to the Colibri. Note that the route of the Monte Olivia/Rio Blanco during March 2010 and 2011 overlap. The maps were generated with Matlab 2016a, the M\_Map 1.7 toolbox (<https://www.eoas.ubc.ca/~rich/map.html>) and using the ETOPO2 Global 2 Arc-minute Ocean Depth and Land Elevation (National Geophysical Data Center/NESDIS/NOAA/U.S. Department of Commerce, 2001. ETOPO2, Global 2 Arc-minute Ocean Depth and Land Elevation from the US National Geophysical Data Center (NGDC). Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory. <http://dx.doi.org/10.5065/D6668B75>. Accessed 1/03/2015).



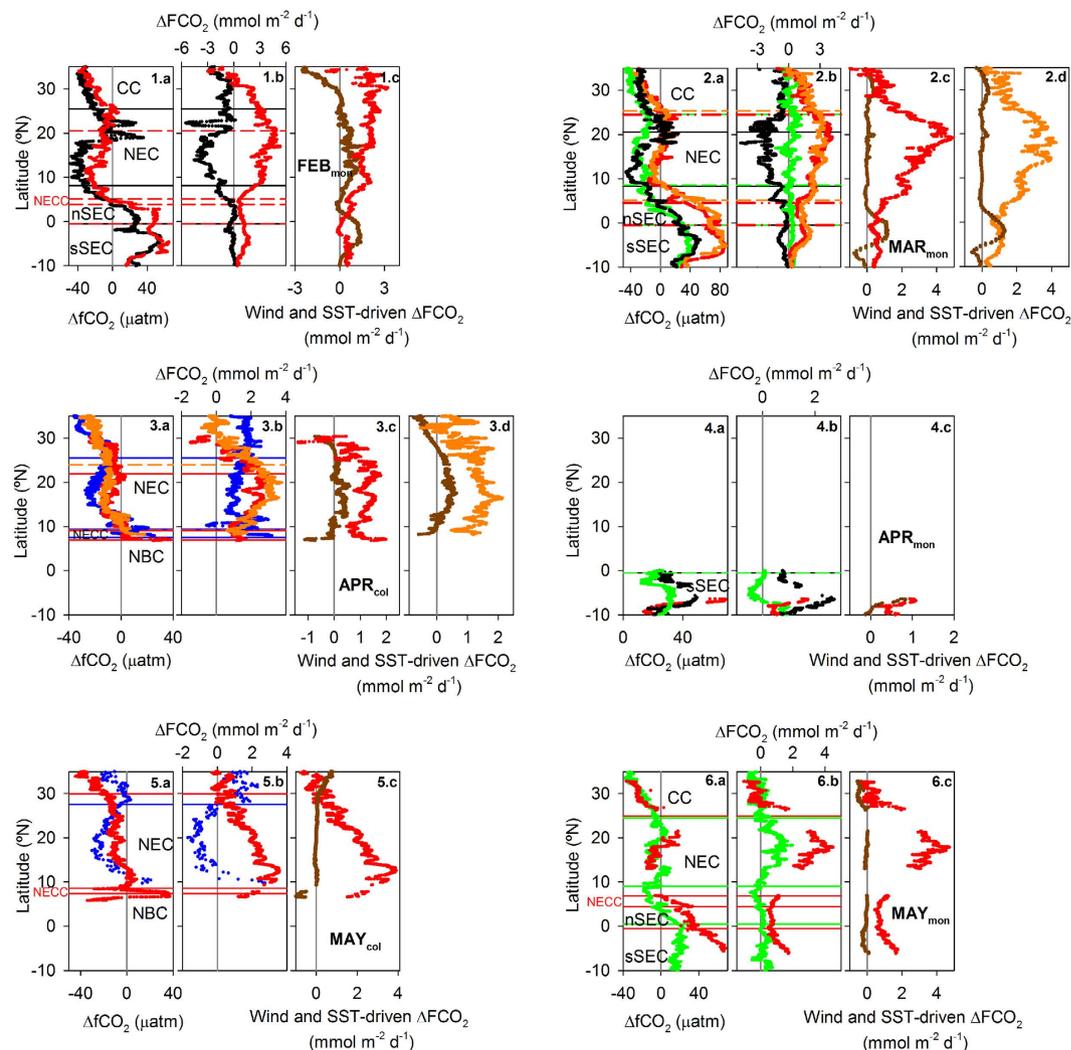
**Figure 3. Schematic view of the main sea surface current system in the tropical and subtropical North Atlantic (red lines).** CC denotes the Canary Current, NEC the North Equatorial Current, NECC the North Equatorial Counter Current, nSEC and sSEC denote the north and south South Equatorial Current respectively and NBC corresponds to the North Brazil Current. The location of the subduction region of the North Atlantic subtropical gyre is also indicated. The climatological sea surface current components, calculated for March for the period 1994–2013 is also shown for reference. These were computed from the Ocean Surface Current Analyses – Real time (OSCAR) data (1° resolution; JPL Physical Oceanography DAAC; developed by ESR). The maps were generated with Matlab 2016a, the M\_Map 1.7 toolbox (<https://www.eoas.ubc.ca/~rich/map.html>) and using the ETOPO2 Global 2 Arc-minute Ocean Depth and Land Elevation (National Geophysical Data Center/NESDIS/NOAA/U.S. Department of Commerce, 2001. ETOPO2, Global 2 Arc-minute Ocean Depth and Land Elevation from the US National Geophysical Data Center (NGDC). Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory. <http://dx.doi.org/10.5065/D6668B75>. Accessed 1/03/2015).

The different water masses transported by the surface current system (Fig. 3) are also clearly reflected in the measured  $f\text{CO}_{2\text{sw}}$ . The equatorial region is dominated by the South Equatorial Current (SEC) that transports Southern Hemisphere  $\text{CO}_2$ -rich waters and the transition between waters originated from both Hemispheres is represented by the different  $f\text{CO}_{2\text{sw}}$  along the Monte Olivia/Rio Blanco tracks at about 4°N (Fig. 1.13 to 1.18).

**Influence of sea surface circulation on SST and sea-air  $\text{CO}_2$  exchange.** Using the Ocean Surface Current Analyses – Real time (OSCAR) data, we estimate the location of the surface currents present along the tracks of the VOS used here (Supplementary Figure S1). From North to South, the large-scale surface currents crossed by the Monte Olivia/Rio Blanco are the CC, the North Equatorial Current (NEC), the North Equatorial Counter Current (NECC) and two branches of the SEC (hereby termed North and South SEC; nSEC and sSEC; Fig. 2 upper panels and 3). Further West, after crossing the subduction region of the North Atlantic subtropical gyre, the Colibri crosses the NEC, the NECC and the North Brazil Current (NBC; Fig. 2 upper panels and 3)<sup>20</sup>.

Excluding the data from the cruises of the Monte Olivia/Rio Blanco apparently affected by upwelling waters in the CC region, we observe significantly higher SST anomalies during 2010 compared to the other years ( $p < 0.05$ ; Supplementary Figure S1) in all the areas delimited by the surface currents but the subduction region of the subtropical gyre (crossed by the Colibri) and the sSEC (crossed by the Monte Olivia/Rio Blanco). There, SST anomalies remain always below 1 °C. Similarly,  $\Delta f\text{CO}_2$  (the difference between  $f\text{CO}_{2\text{sw}}$  and the atmospheric  $f\text{CO}_2$ ,  $f\text{CO}_{2\text{atm}}$ ; positive values denote sea  $\text{CO}_2$  outgassing) is systematically higher during 2010 than during the other years ( $p < 0.0001$ ; Fig. 4, panels labeled a), except in the subtropical gyre. The highest  $\Delta f\text{CO}_2$  differences between the cruises from 2010 and those from other years are found in the NEC ( $18.2 \pm 0.8$ ,  $33.1 \pm 0.8$ ,  $6.2 \pm 0.4$  and  $7.8 \pm 1.2 \mu\text{atm}$  from February to May;  $p < 0.0001$ ), except in May along the Monte Olivia/Rio Blanco track ( $-0.4 \pm 0.6 \mu\text{atm}$ ). The negative  $\Delta f\text{CO}_2$  characteristic of this current during boreal spring is significantly reduced during 2010, with values close to zero and even reversal of the direction of the sea-air  $\text{CO}_2$  exchange during March ( $\Delta f\text{CO}_2$   $3.9 \pm 0.2 \mu\text{atm}$ ,  $n = 2672$ ,  $p < 0.0001$ ; Fig. 4.2.a). In the region of the sSEC,  $\Delta f\text{CO}_2$  during February, March and May 2010 is also significantly higher than those registered during the other years included in this study ( $p < 0.0001$ ; Fig. 4.1.a, 4.2.a and 4.6.a). The abnormal position and intensity of the ITCZ during 2010 explains the increased  $\Delta f\text{CO}_2$  observed in this area<sup>14</sup>.

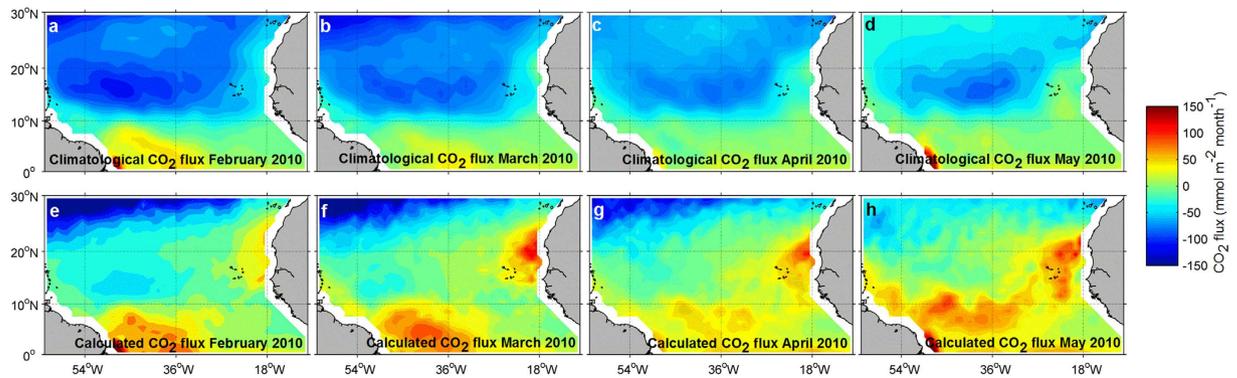
Despite the close longitudinal proximity of the tracks of the two VOS used here in the North of the study area, the significant positive SST anomalies observed in the CC (crossed by the Monte Olivia/Rio Blanco) are not observed in the subduction region of the North Atlantic subtropical gyre (crossed by the Colibri). The CC flows to the South and therefore is not a significant water transport pathway to the subduction region of the subtropical gyre. Furthermore, the sSEC, which transports Southern Hemisphere waters, shows weak SST anomalies during the studied period. At the basin scale, the positive SST anomalies registered from February to May 2010 seem to be generated in the eastern tropical Atlantic, then spreading to west and south following the surface current system. Thus, advection by the surface current system seems to contribute to the spatial distribution of the SST anomalies and  $\Delta f\text{CO}_2$  increase observed in 2010, mainly restricted to the tropical and subtropical North Atlantic.



**Figure 4. Underway  $\Delta f\text{CO}_2$  (shown in panels labeled with a) and  $\text{CO}_2$  flux anomalies ( $\Delta\text{FCO}_2$ ; shown in panels labeled with b).**  $\Delta f\text{CO}_2$  refers to the difference between the underway  $\text{CO}_2$  flux and the normalized  $\text{CO}_2$  flux.  $\Delta\text{FCO}_2$  was calculated as the difference between the underway  $\text{CO}_2$  flux and the normalized  $\text{CO}_2$  flux. The color code denotes the year of each cruise of the Monte Olivia/Rio Blanco (subscript “mon”) and Colibri (subscript “col”) vessels: 2007 (blue), 2009 (black), 2010 (red and orange) and 2011 (green). The contribution of SST and wind anomalies to  $\Delta\text{FCO}_2$  flux during 2010 are also shown in panels labeled with c (and d when more than one voyage of each VOS was performed during that same month in 2010): red and orange dots represent the  $\Delta\text{FCO}_2$  attributed to SST anomalies, while brown dots are the contribution of wind anomalies to the calculated  $\Delta\text{FCO}_2$ . The horizontal lines represent the limits of the surface currents identified in this study for reference.

**Physical contributors to the anomalous sea-air  $\text{CO}_2$  exchange during 2010.** To evaluate the potential contribution of physical parameters to the sea-air  $\text{CO}_2$  flux anomaly of 2010, we compare the underway sea-air  $\text{CO}_2$  flux to that normalized to monthly SST, SSS, wind intensity,  $f\text{CO}_{2\text{atm}}$  and  $f\text{CO}_{2\text{sw}}$ . Excluding the data apparently affected by upwelling waters in the CC and  $\text{SSS} < 35$  (thus excluding the Amazon plume), the normalized  $\text{CO}_2$  flux during 2010 is never significantly higher than that calculated for the other cruises in the area of the CC, the NEC and the NECC ( $p > 0.05$ ; Supplementary Figure S2). In the region of the nSEC and sSEC, the normalized  $\text{CO}_2$  flux during 2010 is still slightly higher than during the remaining cruises (unless during April in the sSEC), which may be related to the uncertainties of the thermodynamic effect of rainfall (ITCZ) over  $f\text{CO}_{2\text{sw}}$ <sup>21</sup>. These results suggest that in the tropical and subtropical North Atlantic, the parameters not accounted in the  $\text{CO}_2$  flux normalization (i.e. primary production,  $f\text{CO}_{2\text{sw}}$  increase due to  $f\text{CO}_{2\text{atm}}$  increase) had little contribution to the  $\text{CO}_2$  flux anomaly observed in 2010.

The strongest positive deviations of the underway  $\text{CO}_2$  flux from the normalized  $\text{CO}_2$  flux are almost systematically registered in the cruises performed during 2010, and mainly concentrated in the Northern Hemisphere (Fig. 4, panels labeled with b). SST and wind intensity anomalies are the most significant parameters contributing to the  $\text{CO}_2$  flux anomalies observed in 2010 (Fig. 4 panels labeled with c and d). SST increase during 2010 explains nearly 100% of the  $\text{CO}_2$  flux anomaly during March and May in the Northern Hemisphere (Fig. 4.2.c, 4.2.d, 4.5.c and 4.6.c; red and orange dots). During February and April, negative wind anomalies act together with positive



**Figure 5.** Basin-scale climatological (a to d) and calculated (e to h) sea-air CO<sub>2</sub> flux during February to May 2010. The climatological sea-air CO<sub>2</sub> flux corresponds to that presented by Landschützer *et al.*<sup>7</sup> and referenced to the year 2010. Based on this referenced sea-air CO<sub>2</sub> flux climatology, the anomalous sea-air CO<sub>2</sub> flux exchange for February to May 2010 was calculated (see discussion). Positive sea-air CO<sub>2</sub> flux denotes sea surface CO<sub>2</sub> outgassing. The maps were generated with Matlab 2016a, the M\_Map 1.7 toolbox (<https://www.eoas.ubc.ca/~rich/map.html>) and using the ETOPO2 Global 2 Arc-minute Ocean Depth and Land Elevation (National Geophysical Data Center/NESDIS/NOAA/U.S. Department of Commerce, 2001. ETOPO2, Global 2 Arc-minute Ocean Depth and Land Elevation from the US National Geophysical Data Center (NGDC) Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory. <http://dx.doi.org/10.5065/D6668B75>. Accessed 1/03/2015).

SST anomalies in explaining the calculated positive CO<sub>2</sub> flux anomalies (Fig. 4.1.c, 4.3.c and 4.3.d; brown dots). Wind intensity determines the gas transfer intensity across the sea-air interface, thus affecting the overall sea-air CO<sub>2</sub> flux. We observe strong negative surface wind anomalies mainly concentrated in the area of the NEC from February to April 2010 (Fig. 1, e to h). These negative wind anomalies lower the magnitude of the sea-air CO<sub>2</sub> exchange and feedbacks the impact of increased SST on reducing the CO<sub>2</sub> absorption capacity in the area of the NEC.

**Basin-scale anomalous sea-air CO<sub>2</sub> exchange in 2010.** SST anomalies in the tropical Atlantic may originate from the zonal or the meridional climate mode. Warming events associated with the tropical Atlantic zonal mode develop under conditions similar to the Pacific El Niño phenomena. Negative wind anomalies in the western equatorial Atlantic precede the reduction of upwelling and the cold tongue in the eastern equatorial Atlantic and overall increased heat content commonly restricted to the equatorial ocean during boreal summer<sup>22</sup>. On the other hand, warming events associated with the Atlantic Meridional Mode (AMM) are characterized by a cross-equatorial SST gradient with positive SST anomalies in the Northern Hemisphere and a Northward shift of the ITCZ, as in boreal spring of 2010<sup>23</sup>. The two modes are interconnected through equatorial waves, where boreal spring, tropical Atlantic warming events associated with positive AMM phases can precede boreal summer, equatorial Atlantic Niño events<sup>24</sup>. The intensity of positive AMM phases are linked and exacerbated by the Pacific El Niño events<sup>23</sup> and positive AMO phases<sup>25</sup>, as observed during 2010<sup>14</sup>. The feedback mechanisms among the AMM, AMO and the El Niño teleconnection mechanisms would thus explain the observed widespread positive SST anomalies in the tropical and subtropical North Atlantic (Fig. 1, a to d). This, in turn, exerts a strong impact over the  $fCO_{2sw}$  and sea-air CO<sub>2</sub> exchange in the tropical and subtropical North Atlantic.

According to the sea-air CO<sub>2</sub> flux climatology calculated from that presented by Landschützer *et al.*<sup>7</sup> and referenced to the year 2010, the region 0°–30°N 62–10°W is a significant atmospheric CO<sub>2</sub> sink during boreal spring (from –8.8 Tg C during February to –4.6 Tg C in May; Fig. 5, a to d). The CO<sub>2</sub> subsaturation of the subtropical North Atlantic largely compensates the net CO<sub>2</sub> efflux characteristic of the equatorial zone (0.3 Tg C during April to 0.8 Tg C during February, from 10°N to the equator). During the warming event of 2010, the net sea-air CO<sub>2</sub> exchange for the 0°–30°N 62–10°W region is 1.6 Tg C of CO<sub>2</sub> outgassing from February to May, contrasting with the high climatological CO<sub>2</sub> absorption for this area during the same period of time (–29.3 Tg C). The strongest sea-air CO<sub>2</sub> exchange anomalies compared to the climatology are concentrated in the region of the NEC, from approximately 25 to 10°N. There, the climatological atmospheric CO<sub>2</sub> uptake of –22.4 Tg C from February to May ceased during 2010 (Fig. 5, e to h). The warm event of this year led to a net CO<sub>2</sub> emission of 1.2 Tg C for these four months, i.e. a difference in the area of the NEC of 23.6 Tg C.

The impact of the Atlantic warm event of 2010 on the sea-air CO<sub>2</sub> exchange is not restricted to the area of the NEC. The weakening of the trade winds during this period promoted also a northward shift of the ITCZ, concomitant with a weaker precipitation in the area, which led to an increase of SSS in the equatorial band and enhanced CO<sub>2</sub> outgassing as shown by Lefèvre *et al.*<sup>14</sup>. Furthermore, the Amazon River presented strong negative discharge anomalies during 2010 which led to a decrease of the atmospheric CO<sub>2</sub> drawdown associated with its plume<sup>26</sup>. Thus, our estimate of 30.9 Tg C of reduced CO<sub>2</sub> uptake in the subtropical and tropical Atlantic (0–30°N, 62–10°W) is a very conservative estimation of the impact of the warming event from February to May 2010. Nevertheless, our estimation is already higher than the overall multidecadal sea-air CO<sub>2</sub> variability reported elsewhere for the entire North Atlantic (20 Tg C)<sup>7–9</sup>. Current surface water warming trends<sup>11</sup> and the increase

in the frequency of extreme climatic events<sup>27</sup> may threaten the mitigation capacity of the North Atlantic over anthropogenic-driven atmospheric CO<sub>2</sub> rise and its climatic consequences. In that regard, global ocean reanalysis products could help in further constrain the effects of climate events as that registered in boreal spring of 2010 over the CO<sub>2</sub> system and the sea-air CO<sub>2</sub> exchange in the tropical Atlantic.

## Materials and Methods

Two automated CO<sub>2</sub> instruments based on infrared detection<sup>28</sup> were installed on board the merchant ships Colibri and Monte Olivia in 2006 and 2008, respectively. The Colibri performs the route Le Havre (mainland France) - Kourou (French Guyana), while the Monte Olivia sails the route Le Havre – Santos (Brazil). In December 2009, the automated CO<sub>2</sub> equipment installed onboard the Monte Olivia was transferred to the Rio Blanco ship performing the same route. Atmospheric and sea surface fCO<sub>2</sub> were determined underway. The ships were also equipped with Seabird thermosalinographs and Druck barometers, thus measuring underway SST, SSS and atmospheric pressure (P). In this study, we used the voyages performed by the two ships from February to May 2010, complemented with those performed during the same months in different years for comparison (Supplementary Table S1). Data collected between 35°N and 10°S were used. A total of 16 voyages recorded data in the selected region and for the selected period (Supplementary Table S1). Due to technical problems, the cruise performed during April 2010 by the Rio Blanco only recorded data from 10°S until 6.4°S. Thus, for the remaining cruises performed during April along the Rio Blanco track, only data recorded from 10°S to the equator are used for comparison.

The atmospheric molar fraction of CO<sub>2</sub> (xCO<sub>2atm</sub>) determined underway the Colibri cruises performed during 2007 and May 2010 were contaminated by the flumes of the ship. Also, during March and April 2011, a problem with the atmospheric pumping onboard the Monte Olivia made xCO<sub>2atm</sub> unavailable. In these cases, the monthly xCO<sub>2atm</sub> recorded at the atmospheric stations of Ascension Island (7.97°S, 14.40°W), Maxaranguape (5.51°S, 35.26°W), Ragged Point (13.17°N, 59.43°W), Tenerife (28.31°N, 16.5°W) and Terceira Island (38.77°N, 27.38°W) of the NOAA/ESRL Global Monitoring Division (<http://www.esrl.noaa.gov/gmd/ccgg/iadv/>) were used. For basin-scale calculations, the monthly xCO<sub>2atm</sub> recorded at Prospect Hill (Bermuda, 32.30°N, 64.77°W) were also used. Monthly measurements performed at these stations were linearly interpolated at the position where underway measurements were performed. Atmospheric fCO<sub>2</sub> (fCO<sub>2atm</sub>) was then calculated according to

$$fCO_{2atm} = xCO_{2atm} (P - p_{H_2O}) C \quad (1)$$

where p<sub>H<sub>2</sub>O</sub> is the water vapor pressure at 100% humidity calculated from SST and SSS and C is the fugacity coefficient calculated from Weiss<sup>29</sup>. The cruise performed by the Colibri during May 2007 lacked P data due to a malfunctioning of the barometer. The monthly atmospheric pressure of the NCEP/NCAR reanalysis project was used in this case. fCO<sub>2atm</sub> derived from the NOAA/ESRL Global Monitoring Division data was also calculated for the remaining ship tracks used in this study for comparison. Good agreement between the fCO<sub>2atm</sub> measured onboard and that obtained from the NOAA/ESRL Global Monitoring Division data was verified, with a statistical difference of 0.76 ± 0.02 μatm among both (paired t-test).

Sea-air CO<sub>2</sub> fluxes (F) were calculated according to

$$F = k S_o (fCO_{2sw} - fCO_{2atm}) \quad (2)$$

where S<sub>o</sub> is the solubility of CO<sub>2</sub><sup>29</sup> and k is the gas transfer velocity. k was calculated according to<sup>30</sup>:

$$k = 0.27 U_{10}^2 (Sc/660)^{-0.5} \quad (3)$$

where Sc is the Schmidt number. U<sub>10</sub> was obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) monthly reanalysis data set (ERA-interim, 0.25° resolution) and interpolated at the position of the underway measurements.

Basin-scale, monthly SST was calculated from the daily NOAA optimum interpolation (OI) SST V2 data (0.25° resolution; NOAA/OAR/ESRL PSD, Boulder, Colorado, USA; <http://www.esrl.noaa.gov/psd/>;<sup>31</sup>) and averaged for each month. Monthly sea surface current velocities were obtained from the Ocean Surface Current Analyses – Real time (OSCAR) data (1° resolution; JPL Physical Oceanography DAAC; developed by ESR) and interpolated at each location where measurements were performed.

The global partial pressure of CO<sub>2</sub> (pCO<sub>2</sub>) climatology presented by Landschützer *et al.*<sup>7</sup> was used for basin-scale calculations of sea-air CO<sub>2</sub> fluxes. This climatology is a neural network-based average of extensive data collected from 1998 until 2011. To reference this climatology to the year 2010, we assumed it corresponds to a climatological situation referenced to the year 2004, the median year of the data they used. Thus, based on their climatology, we first calculated fCO<sub>2sw</sub> from pCO<sub>2</sub> using a climatological SST (see below). We then added the annual increase in fCO<sub>2sw</sub> observed in the oligotrophic North Atlantic (1.1 μatm y<sup>-1</sup>)<sup>32</sup> for the period 2004 to 2010. To obtain an fCO<sub>2atm</sub> referenced to 2010, the basin-scale interpolation of monthly xCO<sub>2atm</sub> was used. fCO<sub>2atm</sub> was then computed using climatological P, SSS and SST. Finally, the climatological sea-air CO<sub>2</sub> exchange for the basin was calculated using a climatological monthly wind speed (see below). Then, we used the thermodynamic dependence of fCO<sub>2sw</sub> with SST<sup>16</sup> to calculate the expected fCO<sub>2sw</sub> during 2010 from the referenced climatological fCO<sub>2sw</sub>. The sea-air CO<sub>2</sub> exchange for the anomalous year 2010 was then calculated with the fCO<sub>2atm</sub>, wind fields, atmospheric pressure, SST and SSS observed in 2010.

Monthly wind, atmospheric pressure (NCEP/NCAR reanalysis project) and SST climatologies were computed for the period 1994–2013. Monthly wind and SST anomalies at the basin scale were then computed for February to May 2010. SST anomalies were also computed along the ship tracks by comparing the measured SST with the interpolated, climatological SST. For basin scale S<sub>o</sub> and p<sub>H<sub>2</sub>O</sub> calculations, SSS data derived from the Soil

Moisture and Ocean Salinity (SMOS) mission and obtained from the Ocean Salinity Expertise Center (CECOS) of the CNES-IFREMER Centre Aval de Traitement des Données (CATDS), at IFREMER, Plouzané (France) were used. The available monthly composites (2010 to 2014) were used to calculate the climatological SSS.

**CO<sub>2</sub> flux normalization and CO<sub>2</sub> flux anomalies calculation.** The different parameters that we considered to potentially contribute to the sea-air CO<sub>2</sub> flux anomaly of 2010 are SST, SSS, wind intensity,  $f\text{CO}_{2\text{atm}}$  and  $f\text{CO}_{2\text{sw}}$ . We interpolated basin-scale climatologies of SST and wind intensity at each position where measurements were taken to obtain the climatological SST and wind intensity for each cruise. We also calculated latitudinal averages of SSS and  $f\text{CO}_{2\text{atm}}$  of monthly cruises of each VOS. Then, we used the thermodynamic dependence of  $f\text{CO}_{2\text{sw}}$  with SSS and SST<sup>16</sup> to calculate the theoretical  $f\text{CO}_{2\text{sw}}$  at these averaged SSS and climatological SST, and thus calculate a normalized CO<sub>2</sub> flux for each cruise together with averaged  $f\text{CO}_{2\text{atm}}$  and climatological wind speed. The CO<sub>2</sub> flux anomaly ( $\Delta F$ ) was then calculated as the difference between the calculated underway CO<sub>2</sub> flux and the normalized CO<sub>2</sub> flux.

To elucidate the relative contribution of the different physical parameters to the CO<sub>2</sub> flux anomaly observed during February to May 2010, we followed the expression presented by Lefèvre *et al.*<sup>14</sup>:

$$\Delta F = \frac{\partial F}{\partial \text{SST}} \Delta \text{SST} + \frac{\partial F}{\partial \text{SSS}} \Delta \text{SSS} + \frac{\partial F}{\partial U_{10}} \Delta U_{10} + \frac{\partial F}{\partial f\text{CO}_{2\text{atm}}} \Delta f\text{CO}_{2\text{atm}} + \frac{\partial F}{\partial f\text{CO}_{2\text{sw}}} \Delta f\text{CO}_{2\text{sw}} \quad (4)$$

where  $U_{10}$  is the wind intensity at 10 m above sea level.

Throughout this study, the errors associated with our results correspond to the standard error of the estimates.

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## Author Contributions

J.S.P.I. and N.L. conceived the idea. N.L. carried out and processed the  $f\text{CO}_2$  measurements. J.S.P.I. analyzed the data and wrote the manuscript with contributions from M.F. and N.L.

## Additional Information

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