1	Changes in the partial pressure of carbon dioxide in the Mauritanian-Cape Verde
2	upwelling region between 2005 and 2012.
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17 ABSTRACT

Coastal upwelling along the eastern margins of major ocean basins represent regions of 18 19 large ecological and economic importance due to the high biological productivity. The 20 role of these regions in the global carbon cycle makes them essential in addressing climate 21 change. The physical forcing of upwelling processes that favor the production in these 22 areas are already being affected by global warming, which will modify the intensity of the upwelling and, consequently, the carbon dioxide cycle. Here, we present monthly high 23 24 resolution surface experimental data for temperature and partial pressure of carbon dioxide in one of the four most important upwelling regions of the planet, the 25 Mauritanian-Cape Verde upwelling region, from 2005 to 2012. This data set provides 26 27 direct evidence of seasonal and interannual changes in the physical and biochemical processes. Specifically, we show an upwelling intensification and an increase of 0.6 Tg a 28 29 year in CO₂ outgassing due to increased wind speed, despite increased primary 30 productivity. This increase in CO₂ outgassing together with the observed decrease in sea 31 surface temperature at the location of the Mauritanian Cape Blanc, 21°N, produced a pH 32 decrease of -0.003 ± 0.001 per year.

34 **1. INTRODUCTION**

35

The excess of CO_2 in the atmosphere, largely responsible for global climate change, has 36 37 prompted research on the role of the oceans in the carbon cycle. The aim in recent decades has been to assess how the oceans act as sources or sinks within the carbon cycle. To 38 39 achieve this goal, hight spatial and temporal observations representative of the 40 distribution of CO₂ fluxes between the ocean and atmosphere are necessary. Automated 41 instruments on volunteer observing ships (VOS) serve to provide as many observations 42 throughtout the global ocean as possible, in addition to the data collected on scientific 43 cruises and at long-term moorings (i.e., Astor et al., 2005: Lüger et al., 2004, 2006; 44 González-Dávila et al., 2005; 2009; Schuster et al., 2009; Ullman et al., 2009; Watson et 45 al., 2009; Padín et al., 2010; Gruber et al., 2002; Dore et al., 2003; Santana-Casiano et al., 46 2007; Bates et al., 2014).

47 With the amount of data already gathered (http://www.socat.info/), climatologies that 48 present average fluxes between the atmosphere and the ocean have been developed, 49 identifying areas acting as a source or sink (Key et al., 2004; Takahashi et al., 2009). However, the low spatial resolution of these databases limits the applicability especially 50 in coastal areas. Upwelling regions are particularly under-represented in such large 51 52 databases. Upwelling presents a dynamic process that raises nutrient and CO₂ rich water 53 from relatively deep areas to the surface. The nutrients reaching the photic zone promote 54 primary production, which consumes CO_2 . This process generates a CO_2 flux into the ocean. On the other hand, the upwelling also brings up CO₂ from deep seawater, which 55 56 generates uncertainty about the actual role of upwelling areas as a source or sink of CO_2 57 (Michaels et al., 2001). Indeed, upwelling areas may act as a source or sink of CO₂ depending on their location (Cai et al., 2006; Chen et al., 2013), where upwelling regions 58

at low latitudes mainly act as a source of CO₂ (Feely et al., 2002; Astor et al., 2005; 59 60 Friederich et al., 2008; Santana-Casiano et al., 2009; González-Dávila et al., 2009) and those at mid-latitudes mainly act as a sink of CO₂ (Frankignoulle and Borges, 2001; Hales 61 62 et al., 2005; Borges et al., 2002; 2005; Santana-Casiano et al., 2009; González-Dávila et al., 2009). Several anthropogenic interactive effects strongly influence Eastern Boundary 63 Upwelling Systems (EBUS), including upper ocean warming, ocean acidification and 64 ocean deoxygenation (Gruber, 2011; Feely et al., 2008; Keeling et al., 2010). Moreover, 65 evidence of increasee wind speed that would favor upwelling (Bakun, 1990; Demarcq, 66 2009; Oerder et al., 2015) supports the possibility of a change in the dynamics of these 67 68 highly productive areas. Recently, eddy-resolving regional ocean models have shown how upwelling intensification can cause a major impact on the system's biological 69 productivity and CO₂ outgassing (Lachkar and Gruber, 2013; Oerder et al., 2015). Wind 70 71 observations and reanalysis products are controversial regarding the Bakun 72 intensification hypothesis (Bakun 1990). Using different wind databases for the Canary 73 region, Barton et al. (2013) concluded that there was no evidence for a general increase 74 in the upwelling intensity off northwest Africa while Marcello et al. (2011) found an 75 intensification of the upwelling system in the same area during a 20-year period while the 76 alongshore wind stress remained almost stable. Cropper et al. (2014) found that coastal 77 summer wind speed increased, resulting in an increase in upwelling-favorable wind speeds north of 20°N and an increase in downwelling-favorable winds south of 20°N. 78 79 Santos et al. (2005; 2012) showed Sea Surface Temperature (SST) was not homogeneous 80 either along latitude or longitude and depending on the upwelling index (UI intensity). Varela et al. (2015) demonstrated opposite results world wide depending on the length of 81 82 data, season evaluated, and selected area within the same wind dataset or between 83 datasets. For the Mauritanian region, when wind stress data were used (Varela et al.,

2015), a more persistent increasing trend in upwelling-favourable winds north of 21°N
and a decreasing trend south of 19°N was determined.

Starting in June 2005, the QUIMA-VOS line visited the Mauritanian-Cape Verde upwelling region northwest of Africa on a monthly basis (Fig. 1 and Supplementary Table S1) producing for the first time a high resolution database of SST and partial pressure of CO_2 expressed as fugacity fCO_2 . This database shows the variations in the CO_2 system under changes in the upwelling conditions in the Canary Ecosystem from 27°N to 10°N for the period 2005 to 2012.

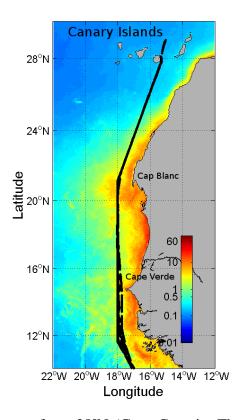


Fig. 1. Ship track in the area from 28°N (Gran Canaria, The Canary Islands) to 10°N (black dots). The locations of Cap Blanc and Cape Verde are indicated. Monthly Ocean Color (oceancolor.gsfc.nasa.gov) data for average chlorophyl *a* concentration (mg m⁻³) were included in a MatlabTM routine and annually averaged. The map has been generated using Matlab 7.12 R2011a.

97

99 **EXPERIMENTAL**

100 **2.1 Study Region.**

The VOS line crosses the East Atlantic Ocean from the north of Europe (English Channel) 101 102 to South Africa, calling at Gran Canaria, the Canary Islands, with a periodicity of two 103 months, which provides monthly data (southward or northward sections). In this work, 104 the area between Gran Canaria at 27°N and 10 °N has been selected in order to study the 105 Mauritanean-Cape Verde upwelling region. In its route south (Fig. 1), the ship leaves 106 Gran Canaria, and goes straight to 100 km off Cap Blanc, at 21°N 17°45'W. It then follows this longitude, passing at 100 km off Cape Verde until 12°N, where it changes 107 direction to Cape Town, reaching 10°N 17°W at 330 km out of the coast of Guinea. 108 109 Between 22°N and 20°N, the ship reaches the 500 m isobath. South of 15°N, the ship 110 moves between 1000 and 500 m isobath. In its route north, the ship follows the same 111 reverse track.

112 2.2 Experimental data

113 Experimental data were obtained under the EU projects Carboocean and Carbochange 114 (www.CarboOcean.org, https://carbochange.b.uib.no/) and now also available at http://www.socat.info/. An autonomous instrument for the determination of the partial 115 116 pressure of CO₂ developed by Craig Neill following NOAA recommendations was 117 installed in a VOS line. This was operated by the MSC company from 2005 to 2008 and 118 the Maersk Company from 2010 to 2012. This VOS line (QUIMA-VOS) run between the 119 UK and Cape Town, from July 2005 to January 2013 (Supplementary Table S1). 120 Temperature was measured at three locations along the sampling circuit: in the intake (SeaBird SBE38L), in the equilibrator (SeaBird thermosalinograph SBE21 and internal 121 PT100 thermometer), and in the oxygen sensor (Optode 3835 AanderaaTM). After the 122

123 seawater pump, the intake is divided in two lines, one feeding the CO_2 system and the 124 other the oxygen sensor, the fluorometer and the seabird thermosalinometer. Differences between equilibrator and intake were constant in time due to the high seawater flow but 125 126 varied among ships due to the different locations of the equipment. Values varied between 127 0.06°C when the equipment was placed close to the intake to 0.35°C, when the equipment was one floor above, inside the engine room. The SST was also obtained from the 128 129 NOAA_OI_SST-V2 data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, 130 USA (http://www.esrl.noaa.gov/psd). These data had a spatial resolution of 1° latitude 131 and 1° longitude and monthly averages were used. The correlation between our experimental SST data and satellite one was better than \pm 1°C, and improved to \pm 0.4°C 132 after removing the most affected upwelling regions (19-22°N and 14-16°N), related to the 133 134 high variability imposed by the upwelling.

135 The CO_2 molar fraction, xCO_2 , in seawater was obtained every 150 s, while atmospheric 136 xCO₂ data were obtained every 180 min. The seawater intake was located at a 10 m depth. The system was calibrated every three hours, by measuring four different standard gases 137 with mixing ratios in the ranges of 0.0, 250-290 ppm, 380-410 ppm and 490-530 ppm of 138 139 CO₂ in the air, provided by NOAA and traceable to the World Meteorological 140 Organisation scale. The precision of the system is greater than 0.5 µatm and the accuracy 141 estimated with respect to the standard gases is of 1 µatm inside the standards range. For xCO₂ values higher than the highest standard (532.04 ppm), the accuracy will be reduced, 142 even when linearity was observed in all cases inside the standards range. The fugacity of 143 144 CO_2 , fCO_2 (µatm), was calculated from xCO_2 after correcting for temperature differences 145 between intake and equilibrator, according to the expressions for the seawater given by 146 DOE (1994). Normalised fCO_2 to the mean SST for the area (T_{mean}) was computed 147 following Takahashi et al. (1993)

148
$$(NfCO_2) = fCO_2 \cdot \exp[0.0423(T_{\text{mean}} - \text{SST})]$$
 (1)

In order to compute a second carbonate system variable, the surface total alkalinity was computed from sea surface salinity (SSS) and SST (Lee et al., 2006). pH_T at the in situ temperature was computed from fCO_2 and A_T and with average annual surface ocean total phosphate and total silicate concentrations of 0.5 and 4.8 µmol kg⁻¹, respectively, from the World Ocean Atlas 2009, using the carbonic acid acidity constants by Merbach et al. (1973) refitted by Dickson and Millero (1987).

155 Air-sea CO₂ fluxes, FCO₂ (mmol $m^{-2} d^{-1}$), were evaluated as

156
$$FCO_2 = 0.24 * k * s * (fCO_2^{sw} - fCO_2^{atm})$$
 (2)

where 0.24 is the scale factor, *k* is the gas transfer velocity, *s* is the CO₂ solubility, fCO_2^{sw} is the seawater fugacity of CO₂ and fCO_2^{atm} is the atmospheric fugacity of CO₂. In order to evaluate ($fCO_2^{sw} - fCO_2^{atm}$), fCO_2^{atm} data were linearly interpolated to the fCO_2^{sw} time vector. A positive value for FCO₂ corresponds with a CO₂ outgassing from the ocean. *k* (cm h⁻¹) was evaluated with the parametrization (Nightingale et al., 2000):

162
$$k = (0.222 * W^2 + 0.333 * w) * (Sc/660)^{-1/2}$$
 (3)

163 where *W* is the wind speed at 10 m above the sea surface (m s⁻¹) and *Sc* is the Schmidt 164 number.

165 The variables involved in estimating FCO₂ data (i.e. fCO_2^{sw} , fCO_2^{atm} , SST and SSS) were 166 fitted to sinusoidal expressions (Lüger et al., 2004) for a given latitude as:

167
$$X(lat)^* = a_0 + a_1(t - 2005) + a_2 sin(2\pi t) + a_3 cos(2\pi t) + a_4 sin(4\pi t) + a_4 sin$$

$$168 \quad a_5 cos(4\pi t) \tag{4}$$

169 where a_i are the fitting coefficients, t is the sampling time expressed as year fraction and X^* represents any of the four fitted variables. This procedure allowed us to re-construct 170 the series of experimental data for periods without monthly data. The variables were 171 decomposed into an interannual term $X(lat)_t^* = a_0 + a_1(t - 2005)$ plus a periodical 172 term $X(lat)_p^* = a_2 sin(2\pi t) + a_3 cos(2\pi t) + a_4 sin(4\pi t) + a_5 cos(4\pi t),$ 173 that is, $X(lat)^* = X(lat)_t^* + X(lat)_p^*$. The periodical term accounts for the high frequency 174 175 seasonal variability, while the interannual one marks the year-to-year trend. First, 176 observations were grouped in a natural year for a given latitude, as if they had been taken 177 in a single year (no correction was done for interannual variability). The mean seasonal 178 climatology data associated with the periodic coefficients (i.e. a₂, a₃, a₄, and a₅) 179 throughout the sampling period were determined. Next, the interannual coefficients a₁ were calculated by fitting the residuals resulting from subtracting the periodical 180 component, $X(lat)_{p}^{*}$, from the original variable X(lat). Fixing these five coefficients (a₁-181 a₅), new distributions for $fCO_2^{sw^*}$, $fCO_2^{atm^*}$, SST^{*} and SSS^{*} were constructed with a daily 182 183 resolution based on the curve fits given for each variable as in Eq. (4), providing the 184 coefficient a₀. The accuracy of this fitting procedure was checked by both computing the correlation between experimental and reconstructed values and by determining the mean 185 186 residuals. The Pearson coefficients were always over 0.87 for SST (average 0.94 ± 0.03), over 0.69 for both fCO_2^{sw} , fCO_2^{atm} (average of 0.79 ± 0.07 and 0.82 ± 0.04, respectively) 187 188 and over 0.67 for SSS (average 0.79 ± 0.07). The mean residual on the determination of 189 those four variables were \pm 3.7 µatm, \pm 1.5 µatm, \pm 0.22 °C, and \pm 0.05 for fCO₂^{sw*}, $fCO_2^{atm^*}$, SST^* and SSS^* , respectively. When the monthly satellite SST values were 190 191 considered, the new SST* function averaged for each month produced values within \pm 192 0.47°C, confirming that this procedure was able to fit non-sampled periods. It was assumed that the same procedure was valid for non-sampled fCO_2 . Finally, daily FCO_2^* 193

time series between 10°N and 27°N with a latitudinal resolution of 0.5° were calculated with a standard error of estimation of 0.5 mmol m⁻² d⁻¹ (15% of error) that produced mean residuals (experimental FCO₂ - FCO₂^{*}) of 0.4 mmol m⁻² d⁻¹ and Pearson correlation coefficients between experimental and computed FCO₂^{*} of r > 0.6, p < 0.01.

198 Chlorophyll-a was calculated from measurements made by the Moderate Resolution 199 Imaging Spectroradiometer (MODIS) aboard NASA's Aqua satellite. We used monthly 200 averages with spatial resolution of 9 km supplied by Ocean Color 201 (oceancolor.gsfc.nasa.gov).

202 Wind data were downloaded from the NCEP CFSR database at 203 http://rda.ucar.edu/pub/cfsr.html developed by NOAA and retrieved from the NOAA National Operational Model Archive and Distribution System and maintained by the 204 205 NOAA National Climatic Data Center. The spatial resolution is approximately $0.3 \times 0.3^{\circ}$ and the temporal resolution is 6 hours. The reference height for the wind data is 10 m. 206

Rainfall data were collected by the Precipitation Radar installed on the Tropical Rainfall Measuring Mission (TRMM) satellite (<u>http://precip.gsfc.nasa.gov</u>). Monthly averages with a spatial resolution of $0.5^{\circ} \times 0.5^{\circ}$ (product 3A12, version 07) were used (Supplementary Fig. S1) in order to explain changes in seasonal surface salinity distributions.

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216 2. RESULTS AND DISCUSSION

217 **3.1 Physical propeties**

The variability of the Mauritanian-Cape Verde upwelling was analyzed in terms of the upwelling index (Nykjaer and Van Camp, 1994) (Fig. 2) using satellite wind data. Negative (positive) UI values correspond to upwelling (downwelling) favorable conditions. The strongest negative values of the index correspond to more intense upwelling. Results clearly distinguish two main subareas in the upwelling system.

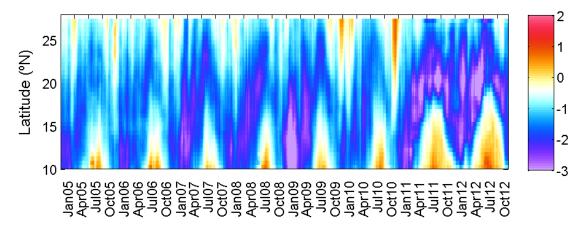


Fig. 2. Time series of upwelling index (UI*10⁻³ m² s⁻¹) in the Mauritanian-Cape Verde upwelling region along the ship track computed following Nykjaer and Van Camp (1994). Cool colours are related to upwelling events and warm colours to downwelling events.

228 1) North of 20°N, the upwelling conditions were favorable throughout the year, although 229 the highest upwellings were observed from March to September with a northward shift 230 from 20° to 22°N. 2) South of 20°N, a marked seasonality was observed with favorable upwelling conditions during autumn and winter, with the maximum intensity observed 231 232 during January and February. In this region, a downwelling regime is present between May and November when the summer trade winds are replaced by the monsoonal winds 233 advecting warm water (Fig. 3a) northward along the shore (Nykjaer and Van Camp, 234 235 1994).

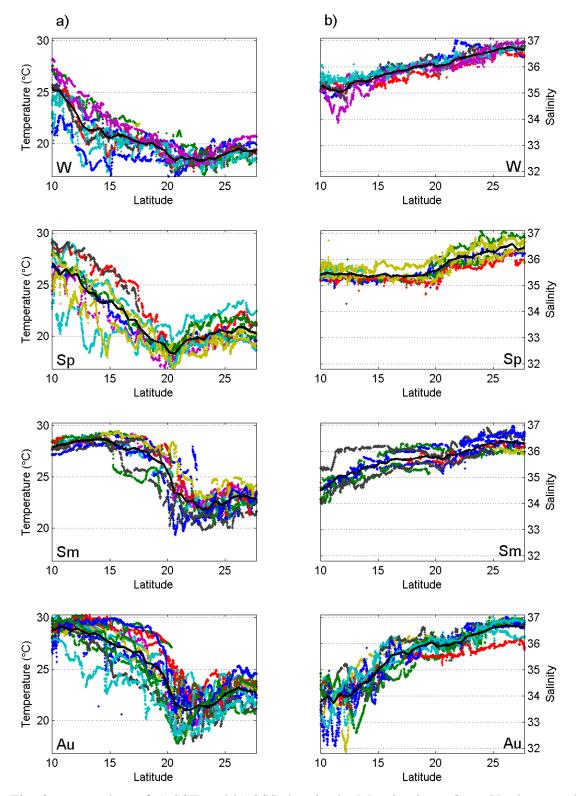


Fig. 3. *In situ* data of a) SST and b) SSS data in the Mauritanian - Cape Verde coastal region grouped by seasons: winter (W, December, January and February), spring (Sp, March, April and May), summer (Sm, June, July and August) and autumn (Au, September, October and November). The averaged values for all cruises in Table S1, are shown in black for each season including the 95% confidence limits. Colour code for each cruise is that indicated in Table S1.

Our results (Fig. 2) are quite consistent with previous research (Nykjaer and Van Camp, 242 1994; Marcello et al., 2011; Santos et al., 2005; 2012; Cropper et al., 2014) but include 243 244 the years 2010 to 2012 where UI at around 20-21°N presented a shift of the upwelling regime intensity from high (-2000 $\text{m}^2 \text{ s}^{-1}$) to strong (-2800 $\text{m}^2 \text{ s}^{-1}$). The analysis of 245 246 upwelling trends along this area has been controversial since it is highly dependent on the selected region (Santos et al., 2012). The inter-annual evolution of UI over the period 247 2005 to 2012 (Fig. 4, green line) for each degree in latitude, iindicates an increase in the 248 UI (mean confidence interval of $9 \text{ m}^2 \text{ s}^{-1}$) as showed by Santos et al. (2012). 249

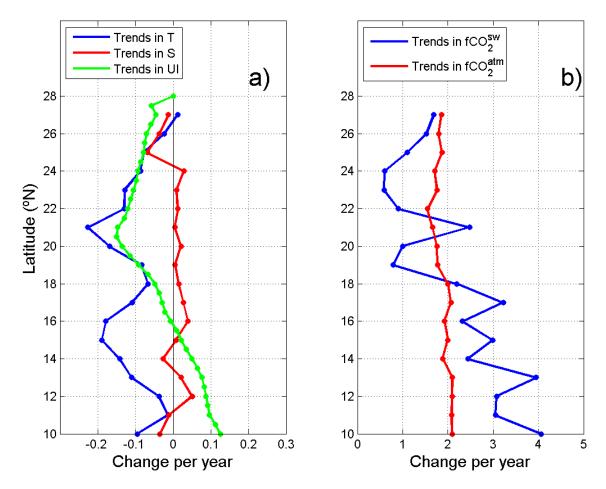


Fig. 4. Latitudinal distribution of the interannual trends for the Upwelling Index (UI*10⁻³) and for the four experimental variables along the QUIMA-VOS line integrated over every degree between 2005 and 2012. The a) panel presents the trends for Upwelling index (UI*10⁻³ m² s⁻¹, mean confidence interval of 9 m² s⁻¹), SST (°C yr⁻¹, confidence interval 0.13°C) and SSS (yr⁻¹, confidence interval 0.06) and the b) panel the trends for fCO_2^{sw} and fCO_2^{atm} (confidence intervals 4.23 µatm and 0.44 µatm).

North of 15°N, the upwelling index confirmed the stronger upwelling observed since 257 1995-1996 in this region after a more than a 10-year (from at least 1982 to 1995) period 258 259 of weaker upwelling (Santos et al., 2012). Local zonal differences between ocean and 260 coastal SST trends determined with satellite data confirmed the intensification of the 261 upwelling regime along the African coast for the period 1982 to 2000 (Santos et al., 2005) extended by Santos et al. (2012) until 2010, and extended in this study until 2012 (data 262 not shown). This has been described as a decadal scale shift of the upwelling regime 263 264 intensity (Marcello et al., 2011; Santos et al., 2012).

South of 15°N, the annual UI values and trends (Fig. 2 and 4) both for the upwelling (values close to -2800 m² s⁻¹ in January) and downwelling (values reaching 1850 m² s⁻¹ in July) periods are becoming stronger. At 11-12N, were downwelling is becoming stronger, this results in negative annual temperature rates that appraches to zero. The UI index serves as an indication of decadal variability of the summer monsoon winds and associated northward advection of warm water along the coast (Santos et al., 2012).

The highest upwelling intensity along the VOS line was located at the capes, Cap Blanc 271 272 and Cape Verde. From satellite chlorophyll-a data, especially off Cap Blanc, giant filaments with chlorophyll concentrations above 1 mg m⁻³ persist year-round, spreading 273 274 from the coast several hundred kilometers offshore (Fig. 1). North of Cap Blanc the 275 upwelled water originates from the North Atlantic Central Water, and mixes with South 276 Atlantic Central Water, SACW, towards the south (Mittelstaedt, 1983). South of Cap 277 Blanc, the upwelling of nutrient rich SACW (Mittelstaedt, 1983) promotes phytoplankton 278 growth between Cap Blanc and Cape Verde. Towards 12°N, upwelling is also fed by the 279 North Equatorial Under Current (Hagen and Schemainda, 1984). Moreover, the entire northwest African coast is also influenced by the African desert dust transport by the mid-280 281 tropospheric Harmattan winds originating from the central Sahara, which supplements

the levels of micronutrients (such as iron) to the adjacent marine ecosystem (Mittelstaedt,
1983; Neuer et al., 2004).

284 The study area is also affected by the migration of the Inter-Tropical Convergence Zone (ITCZ), related to maximum precipitation rates (Hastenrath, 1995). To have a significant 285 286 satellite precipitation record in our region of interest, precipitation data were integrated longitudinally between 25.25°W and 9.75°W. Time series for the latitudinal distribution 287 288 of integrated precipitation (Supplementary Fig. S1) identified the average position of the 289 ITCZ related to maximum precipitation rates. The ITCZ was located at its southernmost position (2°N) during winter, reaching its northernmost position (14-16°N) around 290 291 summer. The ITCZ reached our area of interest (>10°N) from late spring to late summer.

292 The latitudinal distributions of measured SST and SSS along the vessel track are shown 293 in Fig. 3, grouped by seasons. The temperature generally decreased from 10°N to about 294 20°N to 21°N, where the ship meets the Mauritanian upwelling. From there to the north, the temperature rises as the ship leaves the upwelling area on its way to the Canary 295 296 Islands. In situ temperature at 27°N shows temperatures in the range of 18 to 24°C with 297 the minimum in winter and maximum in late summer-early autumn. The annual 298 temperature range was somewhat higher at 20°N, with summer maximum of around 26°C 299 and minimum in spring of about 17°C. At 10°N, temperatures were the highest throughout 300 the year (>25°C), with minimum values in winter and maximum in late spring and late 301 autumn. The low values observed during the end of summer are related to the arrival of 302 the ITZC (Supplementary Fig. S1) at those latitudes. The thermal distribution shows a 303 temperature increase as we move to the Equator and a notable cooling at the upwelled 304 waters off Mauritania. Only during winter time and the begining of the spring, the upwelling of cold water from Cape Verde area was detected. Salinity minimum values 305 306 were normally located at 10°N, increasing to maximum values at the Canaries' latitude. The minimum values of salinity were exceptionally low during autumn from 10°N to 16°N by both the freshwater input from rivers that increase their outflow during this season (Nicholson, 1981) and by the northward shift of the ITCZ during this part of the year.

311 Anomaly fields for temperature and salinity (data not shown) were calculated as the 312 difference between the observations and the mean values at each season for individual 313 latitudes. For temperature, the largest anomalies in winter and spring were located south 314 of 18°N, with values of $\pm 2^{\circ}$ C, related to the seasonal cycle of the Cape Verde upwelling. 315 During summer the pattern changed and the largest anomalies were detected in the 316 upwelling area at 18-22°N, with values of ±5°C when the upwelling index for the 317 Mauritanian area was highest (Fig. 2). In autumn the temperature anomalies were shifted slightly to the north, 20-24°N, with values of ±3°C related to the observed pulses in 318 319 upwelling favorable winds that affected the surface seawater properties. On the other 320 hand, salinity anomalies showed a very homogeneous pattern in all latitudes for winter, 321 spring and summer, with values generally within ± 0.5 . However, during autumn important anomalies south of 18° N were observed, with values in the range of ± 1.5 . In 322 323 this region, the upwelling development, the river discharge and the rainy season 324 controlled the observed distribution (Yoo and Carton, 1990).

The data conclude a permanent annual upwelling regime observed north of 20°N and a seasonal regime across 10–19°N, in accordance with the climatology of previous studies. The data confirm also an increase in upwelling conditions north of 20°N and an increase in downwelling conditions south of 20°N.

329

331 **3.2 Carbon dioxide variability**

The latitudinal distribution of the seasonal fCO_2^{sw} data (Fig. 5a) showed the highest 332 333 values between 18 and 23°N for all seasons due to the variability imposed by the upwelling off Mauritania. fCO_2^{sw} was consintently greater than the fCO_2^{atm} . During 334 winter, when the Cape Verde upwelling develops (Fig. 2), the 12-15°N region also 335 presented higher fCO_2^{sw} values than those in the atmosphere. fCO_2^{sw} data showed a 336 337 latitudinal shift between the seasons following the shift observed in the upwelling index: i.e., in winter, the largest values were located between 19° and 24°N; in spring, they were 338 located between 16° and 22°N; during summer and autumn, the largest fCO2^{sw} values 339 were recorded in the range 20° to 23°N. The difference between fCO_2^{sw} normalized to the 340 341 mean SST of 22°C for the region (N/CO₂^{sw}) and fCO_2^{sw} (ΔfCO_2 , Fig. 5b) reinforced the 342 variability at 20-23°N all year around and at 12-17°N during winter and spring, indicating 343 that upwelling is the major factor contributing to the fCO_2 variability.

According to Takahashi et al. (1993), fCO2^{sw} increases with temperature at a rate of 344 4.3% µatm °C⁻¹ (between 15 and 26 µatm °C⁻¹ in this area) in a thermodynamically 345 controlled system. At 27°N, as SST increases, the rate was only of 7.45 µatm °C⁻¹ due 346 347 mainly to biological uptake and also to the CO₂ outflux. At 20°N the rate became negative with a value of -10.9 μ atm °C⁻¹, clearly indicating the important injection of cool and CO₂ 348 349 rich seawater at the upwelling area. The injection is not being compensated by the solubility nor the biological carbon pumps. At 10°N, the rate was still negative, but only 350 -4.3 µatm °C⁻¹ as a result of the seasonal upwelling. NfCO₂^{sw} was related with SST (data 351 352 not shown) in order to account for effects not removed during normalization. At latitudes 19° to 21°N, in the upwelling vicinity of Cap Blanc, an inverse relationship of 70-353 100 µatm °C⁻¹ was found during winter and spring, while in summer and autumn the 354

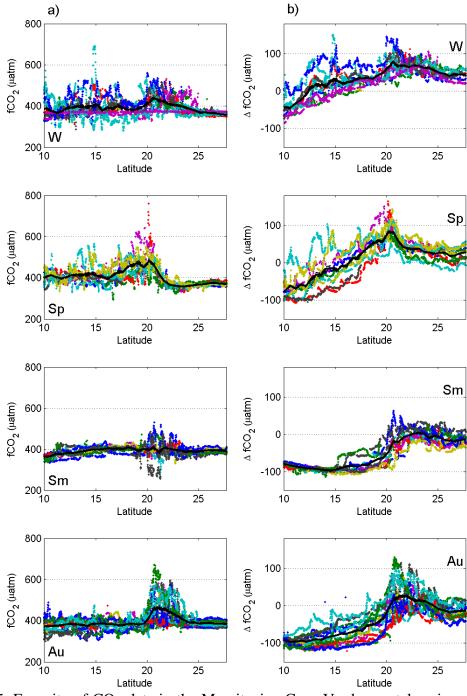


Fig 5. Fugacity of CO₂ data in the Mauritanian-Cape Verde coastal region grouped by seasons: winter (W, December, January and February), spring (Sp, March, April and May), summer (Sm, June, July and August) and autumn (Au, September, October and November). a) fCO_2^{sw} latitudinal distribution. b) Difference between measured and Normalized fCO_2^{sw} values to a constant temperature of 22°C. The averaged values for all cruises in Table S1, are shown in black for each season including the 95% confidence limits. Colour code for each cruise is that indicated in Table S1.

inverse relationship was reduced to 12-18 μ atm °C⁻¹. While the upwelling indexes at those latitudes were quite constant throughout the year, different rates observed should be related to biological consumption of the CO₂ excess. However, during winter and spring the injection of CO₂ in the upwelling is not decreased by the biological activity in the area. But during the Chl-*a* maximum (late spring and summer) most of the CO₂ was consumed and/or exported and, therefore, the rate was strongly reduced.

369 Figure 4 depicts the observed interannual trends (a_1 coefficient in Eq. 4) for the four 370 experimentally recorded detrended parameters, together with the UI trend. Confidence intervals of the computed mean annual values for SST, SSS, fCO2^{atm}, fCO2^{sw} were 0.13°C, 371 372 0.06, 0.44 µatm and 4.23 µatm, respectively. There was a clear SST trend whereby 373 seawater along the VOS line track was getting cooler with maximum cooling rates at the location of Cap Blanc (21°N) and Cape Verde upwellings (15°N) with rates higher than -374 375 0.2°C yr⁻¹. Data from the first three years (2005 to 2008) at 21°N showed lower temperatures with higher cooling rates that reached -0.7°C yr⁻¹, although three years of 376 377 data are not representative. The area crossed by the VOS line along 17°45'W from 22°N to 10°N is located inside the 1000 m isobath that is well inside the mean frontal activity 378 379 in the Canary region, about 200 km wide (Wang et al., 2015). The different changes in 380 temperature in the coastal slope and offshore waters are related to the different origins of 381 the waters upwelled from depths of about 100 m to the surface (Mittelstaedt, 1983) that spread off the coastal area. The offshore water SST is less variable owing to longer 382 383 residence time in the ocean surface. These effects and the fact that the VOS line keeps a 384 track line that crossed the upwelling cells at a distance to the coast that varies among cells, 385 contributes to the observed spatial variability. There was no attempt to compare 386 latitudinal and longitudinal effects on the observed values. Our experimental data, 387 however, does not show any positive SST rates in the upwelling affected area, and only

when the ship approached the Canary Islands, the trends became less negative, reaching a value of $+0.02^{\circ}$ C yr⁻¹ at 27°N, similar to those obtained for oceanic Atlantic water (Bates et al., 2014).

 fCO_2^{atm} for the area presented the interannual increase of about 2 ± 0.3 µatm yrr⁻¹ 391 observed in atmospheric stations, while fCO_2^{sw} presented a heterogeneous distribution. 392 393 South of 18°N the rate of increase was always higher than that in the atmosphere reaching a maximum value of 4.1 ± 0.4 µatm yr⁻¹ at 10°N. At 27°N, fCO₂^{sw} increased at a rate of 394 395 1.7 ± 0.2 µatm yr⁻¹ similar to that determined at the ESTOC time series site (González-Dávila et al., 2010) located at 29°10' N 15°30'W. In the Cap Blanc area, fCO2^{sw} increased 396 at an average rate of $2.5 \pm 0.4 \,\mu$ atm yr⁻¹ with the highest values in the period 2005 to 2008 397 (a rate of 4.6 \pm 0.5 µatm yr⁻¹ was computed with only those years). Around Cap Blanc, 398 fCO_2^{sw} always presented lower rates of increase than in the atmosphere with values well 399 400 below 1 μ atm yr⁻¹. The observed decrease in SST and the trends in fCO_2^{sw} can only be explained by a reinforced upwelling. North of 18°N, the lowest rate of increase in fCO2^{sw} 401 compared to fCO_2^{atm} , together with a decrease in temperature, indicated that upwelling is 402 403 also favoring an increase in the net community production around the Mauritanian 404 upwelling, consuming and/or exporting the CO₂ rich upwelled waters favored by the 405 lateral transport of the Mauritanian current (Lachkar and Gruber, 2013; Varela et al., 406 2015). The upwelling intensification effects observed in the trends of our experimental data support the recent wind stress trends (Crooper et al., 2014; Varela et al., 2015; Santos 407 408 et al., 2012) of increased upwelling-favorable winds, at least for the period 2005-2012 in 409 the Canary upwelling region (Fig. 2 and 4). The intensification of the upwelling results 410 in a change in the measured upwelled water properties due to either higher upwelling 411 velocities or deeper source upwelled waters. However, what remains unclear from these 412 records is to what extent those changes reflect upwelling variations due to climate change 413 forcing versus natural decadal variability in the upwelling areas occurring over414 interannual timescales.

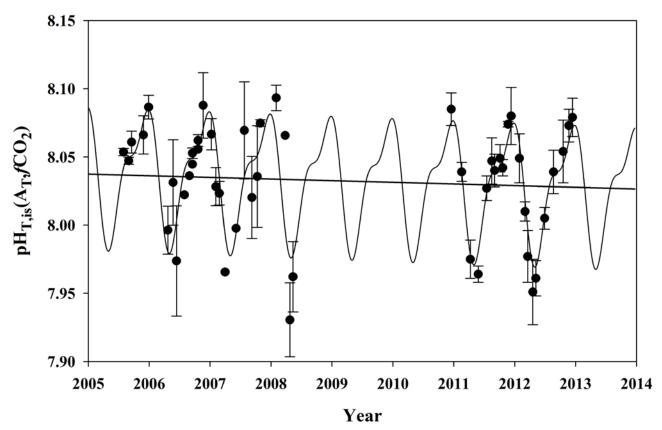


Fig. 6. pH at *in situ* SST in total proton scale computed from total alkalinity (based on regional correlations with SST and SSS, Lee et al., 2006) and fCO_2 at 21 ± 0.25 °N. The error bar represents the standard deviation of the computed data for each cruise for the selected latitude. The black line shows the harmonic fitting Eq. (4) for the data and the corresponding linear trend.

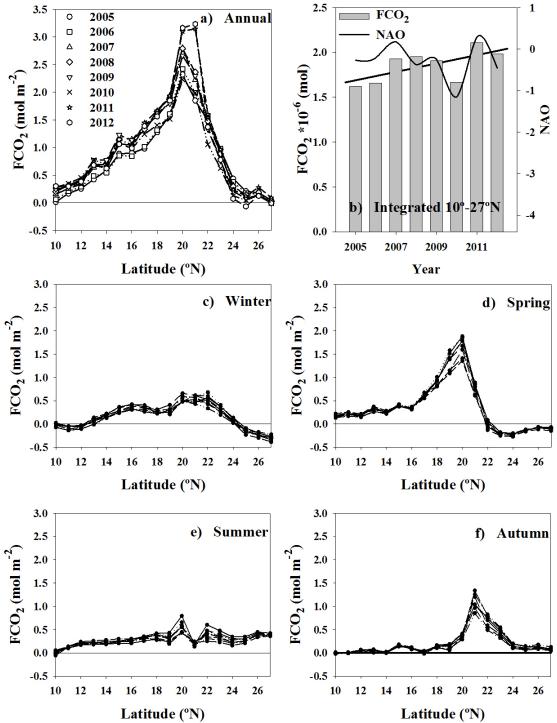
Because the upwelling intensity is changing, other variables will also be affected. pH_{T,is} at 21 \pm 0.25°N was computed from *f*CO₂ and alkalinity pairs of data. Alkalinity was computed from regional correlations with SST and SSS (Lee et al., 2006) which could under-represent seasonal and interannual variations in upwelling areas. However, pH computed from *f*CO₂ values are relatively insensitive to errors in A_T, and *f*CO₂ controls the magnitude and variability of pH (a 60 µmol kg⁻¹ change in A_T will affect a 0.1% in pH, that is, about 0.01 pH units). Figure 6 depicts the computed pH_{T,is}(A_T, *f*CO₂) data

and the harmonic fitting Eq. (4) providing seasonal variability and interannual trend. 428 Considering the small systematic biases in interannual dynamics, we determined a 429 430 decrease in pH at a rate of -0.003 ± 0.001 per year (Fig. 6). This decrease is one of the highest rate values determined in several time series stations (Bates et al., 2014), where 431 432 oceanic SST has only slightly increased in the last decades. However, at the Mauritanian 433 upwelling area and at the location where our VOS line approached this region, SST decreased at a rate of $-0.22 \pm 0.06^{\circ}$ C yr⁻¹ (Fig. 4). Solely, this decrease in temperature 434 would increase the pH by a rate of +0.004 yr⁻¹ and the fCO₂ would decrease by 4 µatm yr⁻¹ 435 ¹. The net effect of the increase in the amount of rich CO₂/low pH upwelled waters in the 436 Mauritanian upwelling would be, therefore, a decrease in the pH of over -0.007±0.002 437 units yr⁻¹ and an increase in fCO_2 of +6.5 ± 0.7 µatm yr⁻¹ (with periods where those rates 438 could reach values of 0.015 yr⁻¹ in pH and 10.5 μ atm y⁻¹ in fCO₂ as recorded during 2005-439 440 2008). Those values are greatly compensated by the important decrease in the SST resulting in the determined rates of -0.003 ± 0.001 pH units and $+2.5 \pm 0.4$ µatm of fCO₂ 441 442 per year.

This new data set of experimental values confirmed a decrease in SST and trends in fCO_2^{sw} than can only be explained by a reinforced upwelling conditions, that favor an increase in the net community production around the Mauritanian upwelling together with a more corrosive environment with pH values that decrease by over -0.007±0.002 units at 21°N. However, the decrease in SST in the upwelling cell buffers this rate to values around -0.003 ± 0.001 pH units yr⁻¹ and +2.5 ± 0.4 µatm yr⁻¹ in fCO_2 , still among the highest observed in other time series.

450

451 **3.3 Fluxes of CO**₂



452 Fig. 7. Latitudinal distribution of seasonal and annual CO_2 fluxes, FCO_2 (mol m⁻²). Fluxes of CO₂ were computed using Nightingale et al. (2000) parametrization and 453 454 satellite winds with a resolution of 6 hours. a) Integrated year-to-year from 2005 to 2012 and b) latitudinally integrated for 2005 to 2012 together with annual values for NAO 455 456 index. Latitudinal distribution of FCO₂ seasonally integrated from 2005 to 2012 are depicted for winter (c, December, January and February), spring (d, March, April and 457 458 May), summer (e, June, July and August) and autumn (f, September, October and 459 November).

461 The annual air-sea CO₂ flux for the full domain was positive (Fig. 7a), with the area off with values close to 3.3 mol CO₂ m⁻² (Fig. 7a). North of 24°N, in the area not affected by 462 the coastal upwelling, an average flux of $+0.14 \pm 0.03$ mol CO₂ m⁻² was determined. The 463 ingassing observed during winter and spring of -0.16 ± 0.03 mol CO₂ m⁻² for the full 464 period (Fig. 7) was surpassed by the outgassing during summer and autumn of 0.28 \pm 465 0.14 mol CO₂ m⁻². South of 24°N, it was observed that during spring (Fig. 7d) the 466 photosynthetic activity was not intense enough to uptake the CO₂ injected by the strongest 467 upwelling in the surface waters and thus the area acted as a source of CO₂ with values 468 reaching 1.9 mol CO₂ m⁻² in 2012. During summer (Fig. 7e), primary producers and 469 470 lateral advection of warm waters by the Mauritanian current could consume/export the CO₂ rich waters reaching values of 0.5 mol m⁻². During autumn (Fig. 7f), only the area 471 472 between 20°N and 23°N acted as a source of 1-1.5 mol CO₂ m⁻², while the rest was almost in equilibrium. Late autumn-winter upwelling in the 14° to 17°N region contributed to an 473 increased outgassing with a second annual submaximum of about 0.4 mol CO_2 m⁻² in 474 winter (Fig. 7c). South of 14°N, annual CO₂ fluxes decreased from about 0.7 mol m⁻² at 475 14°N to being roughly in equilibrium at 10°N. 476

477 The integrated CO₂ fluxes for the area 10°N to 27°N along the VOS line section for the vears 2005 to 2012 (Fig. 7b) were between 1.6 and 2.1 10⁶ mol, with an important annual 478 variability. FCO₂ increased during the studied period by $0.05 \pm 0.02 \cdot 10^6$ mol yr⁻¹. The 479 augment in FCO₂ is related to the observed increase in wind speed (Fig. 4, indicated as 480 481 UI) north of 16°N. North of 19°N, the influence of wind speed far surpassed the effect of the smaller annual rate of increase in fCO_2^{sw} relative to fCO_2^{atm} , with an exception at 482 21°N (Fig. 4). South of 16°N, the decrease in wind speed did not exceed the effect of the 483 incremental change in $(fCO_2^{sw} - fCO_2^{atm})$ associated with the increased downwelling 484

indexes (Fig. 4; Santos et al., 2012), resulting in a slightly increasing FCO₂. The 485 486 variability observed in the annual integrated CO₂ fluxes (Fig. 7b) was related with the 487 basin-scale oscillations, the North Atlantic Oscillation (NAO) index and the East-Atlantic 488 Pattern (EA) (http://www.cpc.ncep.noaa.Gov/data/teledoc/ telecontents.shtml). Cropper 489 et al. (2014) found winter upwelling variability was strongly correlated with the winter 490 NAO (r values ranged from 0.50 at 12–19°N to 0.59 at 21–26°N), due to the influence of 491 the Azores semi-permanent high-pressure system on the strength of the trade winds. The 492 annual integrated FCO₂ was related with the annual NAO index (Fig. 7b) with a similar 493 r = 0.54, even when fluxes are not only controlled by wind strength. However, Fig. 7a 494 clearly indicates that the Mauritanian upwelling area was the most important contributor 495 to FCO_2 in the study area. The FCO_2 was not significantly correlated with the winter 496 NAO (r = 0.23). Also, the EA index, which represents a southward-shifted NAO-like 497 oscillation, presented a lower significant value (r = 0.48) (trends not shown), in agreement 498 with the upwelling index (Cropper et al., 2014). Overall, the correlation between fluxes 499 and climate indexes describing the main mode of variability across the Atlantic sector 500 may be directly related to the Azores High and its influence on the trade wind strength.

501 FCO₂ values along the QUIMA-VOS line were used in order to compute a flux budget 502 for the Mauritanean-Cape Verde region. The observed values were assumed to be valid 503 for at least 100 km to both sides of the QUIMA-VOS line. In this case, the total flux of CO₂ being ejected to the atmosphere would reach a value of 16 Tg of carbon dioxide a 504 year for the period 2005-2012, with a rate of increase of 0.6 Tg yr⁻¹. However, it should 505 506 be considered that the export of the rich fCO_2 upwelled water with high nutrient 507 concentration off the coastal areas would promote a decrease in surface fCO₂ values 508 during productive seasons (as those observed north and south 21°N) that will result in an

ingassing of CO₂. This could balance the observed outgassing increase in a more globalscale.

511 **4. CONCLUSIONS**

512 The Mauritanian-Cape Verde upwelling area's sensitivity to climatic forcing on 513 upwelling processes strongly affects the CO_2 surface distribution, ocean acidification 514 rates and air-sea CO_2 exchange.

515 The experimental SST and carbon dioxide system variables results for the period 2005 to 516 2012 confirm upwelling intensification at the Mauritanian-Cape Verde upwelling system. 517 Furthermore, we have shown that upwelling regions at low-mid latitudes are important 518 sources of CO₂ to the atmosphere. As a direct result, the pH is decreasing at a rate of -519 0.003 ± 0.001 per year. Importantly, the amount of emitted CO₂ is increasing annualy at 520 a rate of 0.6 Tg due to stronger wind stress, even when primary production seems to also 521 be enhanced in the upwelling area. The montly record in this EBUS is not yet long enough 522 to determine the extent to which these changes can be attributed to natural decadal 523 variability. These VOS line must be maintained for years to come, and will continue to 524 be on of the most significat contributors to our knowledge of how ocean surface waters 525 are being affected by present and future climate change. The results from VOS lines can 526 provide accurate data for changes in SST, FCO₂ and, consequently, upwelling 527 intensification effects due to global change conditions under decadal natural variability.

529 Data availability.

- 530 All data are free available at the SOCAT data base, <u>http://www.socat.info/</u> and at the
- 531 Carboocean and Carbochange web pages www.CarboOcean.org,
- 532 <u>https://carbochange.b.uib.no/</u>, respectively
- 533
- 534

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543 Author contributions

- 544 M.G.D. and J.M.S.C worked in the equipment installation, data collection and designed
- 545 the study. F.M. processed the data, generated figures and results. All of them collaborated
- 546 in the discussion of the data and the writing of the paper.
- 547

548 **Competing interests**

- 549 There is not any competing interest.
- 550 551
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725 LEGEND FOR FIGURES

Fig. 1. Ship track in the area from 28°N (Gran Canaria, The Canary Islands) to 10°N
(black dots). The locations of Cap Blanc and Cape Verde are indicated. Monthly Ocean
Color (oceancolor.gsfc.nasa.gov) data for average chlorophyl *a* concentration (mg m⁻³)
were included in a MatlabTM routine and annually averaged. The map has been generated
using Matlab 7.12 R2011a.

Fig. 2. Time series of upwelling index (UI $*10^{-3}$ m² s⁻¹) in the Mauritanian-Cape Verde upwelling region along the ship track computed following Nykjaer and Van Camp (1994). Cool colours are related to upwelling events and warm colours to downwelling events.

Fig. 3. *In situ* data of a) SST and b) SSS data in the Mauritanian - Cape Verde coastal region grouped by seasons: winter (W, December, January and February), spring (Sp, March, April and May), summer (Sm, June, July and August) and autumn (Au, September, October and November). The averaged values for all cruises in Table S1, are shown in black for each season including the 95% confidence limits. Colour code for each cruise is that indicated in Table S1.

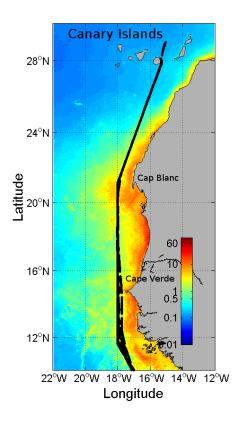
Fig. 4. Latitudinal distribution of the interannual trends for the Upwelling Index (UI*10⁻³) and for the four experimental variables along the QUIMA-VOS line integrated over every degree between 2005 and 2012. The a) panel presents the trends for Upwelling index (UI*10⁻³ m² s⁻¹, mean confidence interval of 9 m² s⁻¹), SST (°C yr⁻¹, confidence interval 0.13°C) and SSS (yr⁻¹, confidence interval 0.06) and the b) panel the trends for fCO_2^{sw} and fCO_2^{atm} (confidence intervals 4.23 µatm and 0.44 µatm).

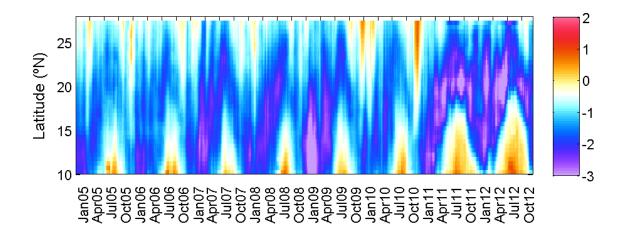
Fig 5. Fugacity of CO₂ data in the Mauritanian-Cape Verde coastal region grouped by
seasons: winter (W, December, January and February), spring (Sp, March, April and
May), summer (Sm, June, July and August) and autumn (Au, September, October and

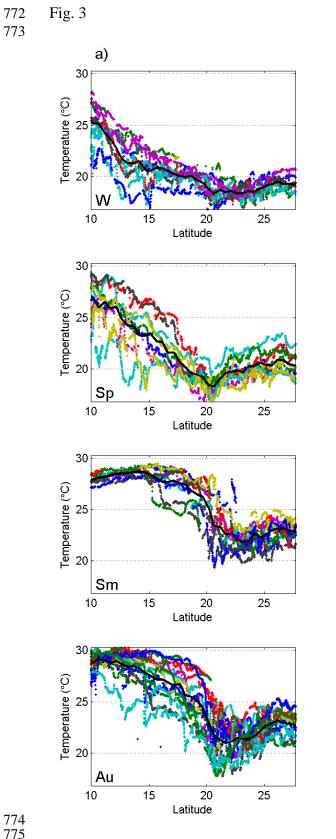
November). a) fCO_2^{sw} latitudinal distribution. b) Difference between measured and Normalized fCO_2^{sw} values to a constant temperature of 22°C. The averaged values for all cruises in Table S1, are shown in black for each season including the 95% confidence limits. Colour code for each cruise is that indicated in Table S1.

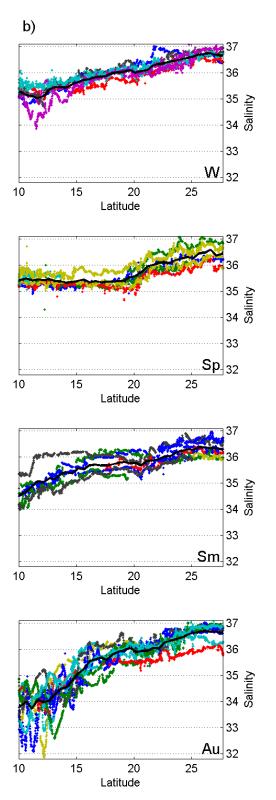
Fig. 6. pH at *in situ* SST in total proton scale computed from total alkalinity (based on regional correlations with SST and SSS, Lee et al., 2006) and fCO_2 at 21 ± 0.25 °N. The error bar represents the standard deviation of the computed data for each cruise for the selected latitude. The black line shows the harmonic fitting Eq. (4) for the data and the corresponding linear trend.

Fig. 7. Latitudinal distribution of seasonal and annual CO_2 fluxes, FCO_2 (mol m⁻²). 759 760 Fluxes of CO₂ were computed using Nightingale et al. (2000) parametrization and 761 satellite winds with a resolution of 6 hours. a) Integrated year-to-year from 2005 to 2012 762 and b) latitudinally integrated for 2005 to 2012 together with annual values for NAO 763 index. Latitudinal distribution of FCO₂ seasonally integrated from 2005 to 2012 are 764 depicted for winter (c, December, January and February), spring (d, March, April and 765 May), summer (e, June, July and August) and autumn (f, September, October and 766 November).

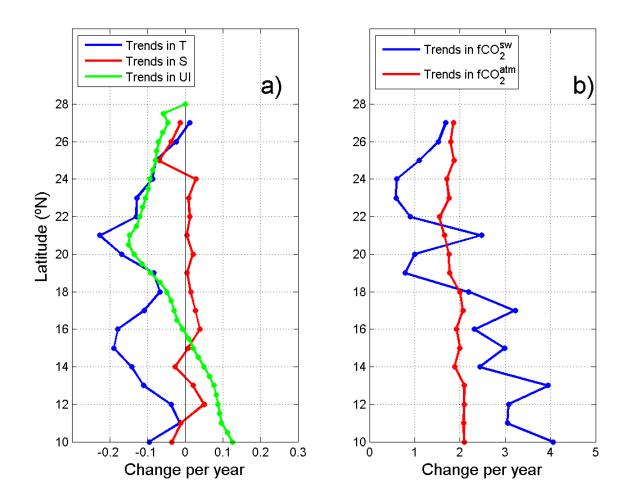














780 Fig. 5

