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Observed trends of pCO_2 and air-sea CO_2 fluxes in the North Atlantic Ocean

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Abstract Observed partial pressure of carbon dioxide (pCO_2) and temperature data in surface and mixed layer seawater of the Northeast (49°N, 16.5°W) and Northwest (56.5°N, 52.6°W) Atlantic Ocean time series sites have been analyzed for seasonal variability and air-sea CO_2 fluxes. The NE PAP data showed an annual mean pCO_2 of 335.9 ± 89.6 µatm (2003), 286.7 ± 103.5 µatm (2004), and 335.9 ± 89.6 µatm (2005). The annual data for NW KI deployments indicated annual pCO_2 average of 336.6 ± 14.3 and 359.1 ± 25.3 µatm for 2004 and 2005 respectively. The oceanic pCO_2 distribution across the spatial gradients over a seasonal timescale is relatively homogeneous with marked seasonal variability. These data indicated consistently the undersaturation of oceanic surface water at the sites and thus a perennial carbon sink. Sea surface pCO_2 trend is marked by summertime minimum and wintertime maximum, while depicting anti-phase patterns with the observed temperature signals. Seasonal to annual CO_2 fluxes indicated a year-round CO_2 invasion of the NE and NW basins. Estimated net basin-scale CO_2 uptake fluxes of 2.96 ± 1.73 and 1.84 ± 1.3 mol m^{-2} CO_2 a^{-1} were obtained for NE PAP (2nd - 4th) and NW K1 deployments, respectively.

Keywords pCO_2; air-sea CO_2 fluxes; seasonal variability; temperature trends; North Atlantic Ocean

Introduction

The North Atlantic Ocean is regarded as the largest ocean sink for atmospheric carbon dioxide (CO_2), based both on observational estimates (Schneider et al., 1992; Kuss et al., 2006; Takahashi et al., 2002; 2009), and forward and inverse modeling results (Gloor et al., 2003; McKinley et al., 2004; 2008). The strong CO_2 sink capacity of the North Atlantic Ocean is largely attributed to two principal factors vis-a-vis the counteractive effect of vertical circulation in which large volumes of surface water driven poleward by strong currents, cools and absorbs huge quantities of atmospheric CO_2 before getting sunk to mixed layer depth during the wintertime, and also to the effective and sustained carbon and nutrients (phosphorus, silica and iron) uptake (Takahashi et al., 2002, 2009; Schuster and Watson, 2007; Körtzinger et al., 2008a). However, observations have indicated substantial variability in the uptake of CO_2 spatially (Watson et al., 1991) and temporally (Gruber et al., 2002).

The exchange of CO_2 between the ocean and the atmosphere is a major biogeochemical process that regulates the fate and rate of increase of anthropogenic CO_2, which will in turn determine the rate of likely climate change. This biogeochemical process is significantly controlled by prevailing pCO_2 existing between the atmosphere and surface of the ocean. Sea surface pCO_2 is governed by physical and biological factors such as change of sea surface temperature (SST) (Schuster et al., 2009), deep convective mixing, re-stratification (Straneo, 2006), entrainment of CO_2 enriched deeper water (Avsic et al., 2006; Körtzinger et al., 2008b), salinity (Dickson et al., 2002), phase of the North Atlantic Oscillation (Thomas et al., 2008), consumption by marine biota linked to the availability of surface nutrients (productivity / respiration) (Behrenfeld et al., 2006). The net CO_2 transfer is a function of the difference in the pCO_2 at the air-sea interface, and of the exchange processes in the atmosphere and the ocean (Takahashi et al., 2009; Omstedt et al., 2009).

There is a growing understanding that adequate parameterization of CO_2 flux is a significant factor in the quantification of spatially resolved air-sea CO_2 exchange (Rutgersson et al., 2008; Takahashi et al., 2009). The assessment is crucial for climate modeling

owing to the fact that CO_2 is the major driver of anthropogenic climate change. Additionally, in order to understand how the changing global environment may alter the carbon cycle, it is necessary to further analyse the fluxes and examine the physicochemical and biological processes that determine them.

1 Data and methods

1.1 *Sources of observed data*

The observed data used for this research where obtained from the Porcupine Abyssal Plain and K1 Central Labrador Sea oceanographic mooring sites in the North Atlantic Ocean (Figure 1).

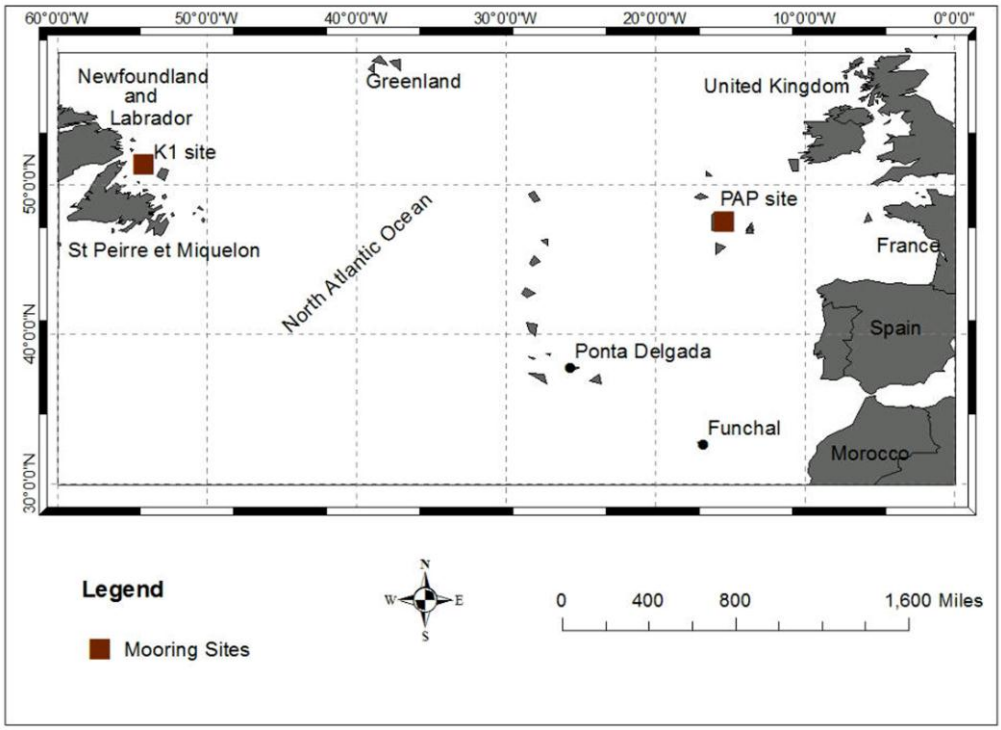

Figure 1 Map of the North Atlantic Ocean showing the Porcupine Abyssal Plain (49°N, 16.5°W) and Labrador Sea (56.5°N, 52.5°W)

1.2 *Air-sea CO_2 fluxes estimation*

The calculation of CO_2 flux (fCO_2) in ocean and climate models is based on the indirect bulk method. The net exchange of CO_2 (f) between the ocean surface and the atmosphere is estimated from the air-sea difference in partial pressure of CO_2 (ΔpCO_2) and the gas transfer velocity (k) using the equation (Wanninkhof, 1992, 2007; Donelan and Wanninkhof, 2002):

$$fCO_2 = k \times (\upsilon)K_0 \times \Delta pCO_2 \qquad (1)$$

Where k is the gas transfer velocity of CO_2 exchange, υ is the wind speed, K_0 is the solubility of CO_2 in seawater and is a function of salinity and temperature (Weiss, 1974), and ΔpCO_2 is mean air-sea pCO_2 difference,

$$\Delta pCO_2 = [(pCO_2\text{-air}) - (pCO_2\text{-sw})] \qquad (2)$$

Where pCO_2-air and pCO_2-sw represent the respective partial pressure of carbon dioxide in the atmosphere and seawater (Rutgersson et al., 2008).

The transfer velocity, k, is regarded as a function of wind speed, υ, and the Schmidt number (Sc), although this is still controversial (Weiss et al., 2007; Rutgersson et al., 2008). The Schmidt number (Sc) is the ratio of the kinematic viscosity of seawater to the diffusion coefficient of the considered gas. For wind speeds larger than 5 ms^{-1}, k is proportional to $Sc^{-1/2}$ (Liss and Merlivat, 1986). Different functions which refer to Sc = 660 (CO_2 at 20 C) have been proposed to describe k_{660} as a function of wind speed at a reference height of 10 m (υ_{10}). Wanninkhof (1992) suggested a quadratic equation:

$$k_{660} = 0.31\,\upsilon^2_{10} \qquad (3)$$

which gives k for any other Sc as

$$k = 0.31\,\upsilon^2_{10}\ \sqrt{660/Sc} \qquad (4)$$

A cubic dependence is given from Wanninkhof and McGillis (1999):

$$k = 0.0283\ \upsilon^3_{10}\ \sqrt{660/Sc} \quad (5)$$

Moreover, recent investigation evaluating the transfer velocity-wind speed relationship using a long-term series of direct eddy correlation CO_2 flux measurements from the Baltic Sea suggests a combination of quadratic and linear wind speed dependence (Weiss et al., 2007; Rutgersson et al., 2008).

$$k = (0.365\ \upsilon^2_{10} + 0.46\ \upsilon^2_{10})\sqrt{660/Sc} \quad (6)$$

A positive flux (fCO_2) value represents a net CO_2 exchange from sea to the atmosphere and a negative flux value refers to the net CO_2 exchange from the atmosphere to the sea. For the purposes of this research, 8-day averages of air-sea pCO_2 difference (ΔpCO_2), SST, wind speed at 10 m height, and mixed layer depth are produced, and calculations of 8-day CO_2 flux densities, *f*, were performed using equation (4).

2 Results and discussion

2.1 *Seasonal to monthly pCO_2 net flux in North Atlantic NW / NE basins*

The emerging trend of the seasonal pCO_2 cycle at the PAP observatory site indicates high pCO_2 difference between the seawater relative to atmospheric pCO_2, showing a persistent undersaturation of surface waters by ΔpCO_2 of about 70 µatm in summer of 2004 (Figure 2). This predicts an influx of CO_2 from the atmosphere into the ocean, and the trend notably followed an increasing seawater surface temperature (warming). Winter deep convection has been established as a mechanistic process of exposing CO_2-enriched subsurface water to the seawater surface (McKinley et al., 2004b). This mechanism is markedly observed during the winter months (Figure 2) where relatively low or damped ΔpCO_2 were obtained.

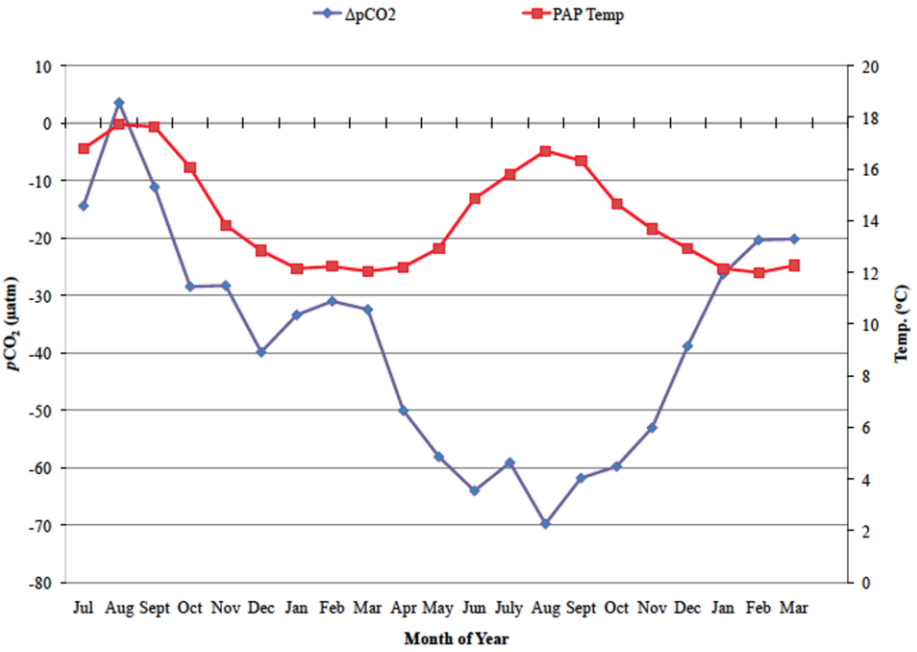

Figure 2 Monthly ΔpCO_2 between July 2003 – March 2005 plotted as a function of SST at the PAP observatory

In other words, the time-trend ΔpCO_2 flux variability indicates relatively damped ΔpCO_2 during wintertime and enhanced ΔpCO_2 in summertime. Also, uptake of anthropogenic CO_2 from the atmosphere increases during wintertime resulting in relatively low ΔpCO_2. However, a positive ΔpCO_2 was observed in early summer of 2003 (August 2003) resulting in a possible efflux of CO_2 into the atmosphere leading to a decrease in net ocean CO_2 uptake during the summertime. This could be attributed to a dominant DIC-driven pCO_2.

Considering the average monthly CO_2 flux variability at the NW K1 Central Labrador Sea, a relatively high sea-air ΔpCO_2 of about 60 µatm was recorded in September 2004 as a negative CO_2 flux (Figure 3). However, decreasing CO_2 sink of the time series location is observed following seasonal change from summertime of 2004 to wintertime. Uptake of CO_2 by the ocean almost equilibrated with the atmospheric pCO_2 during the peak period of the winter months, thereby creating a near saturated condition with relatively low sea surface temperature. Evidently,

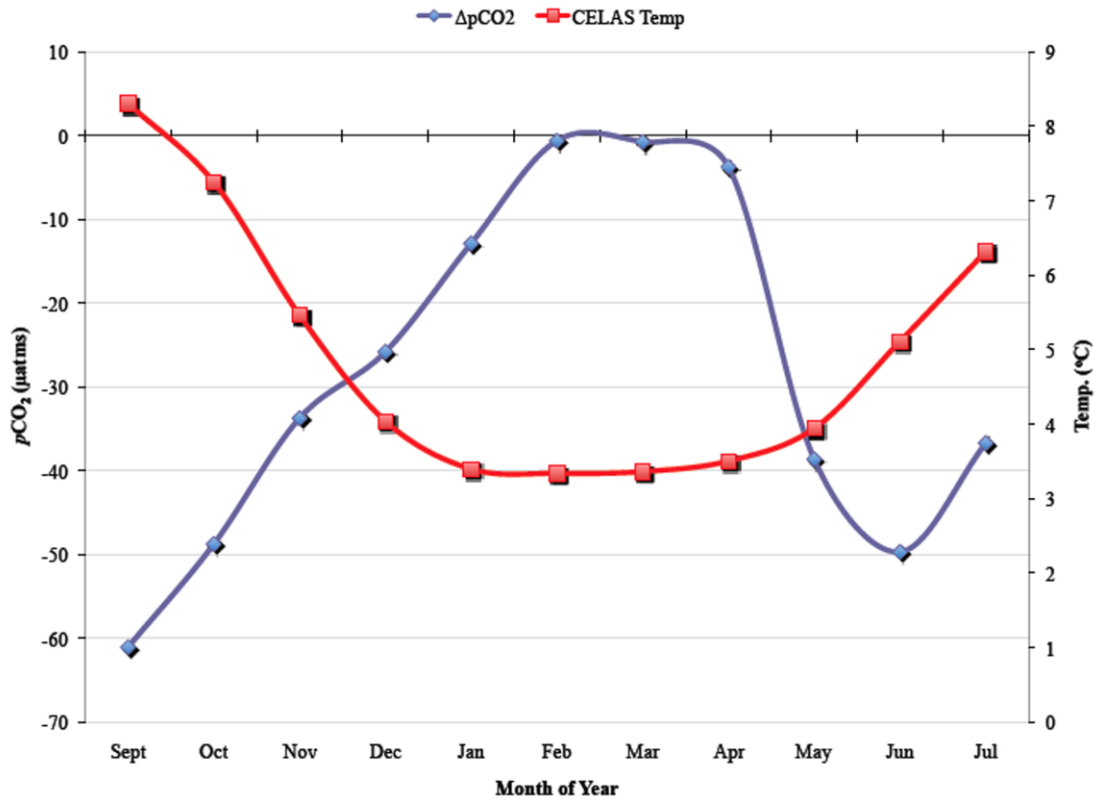

Figure 3 Monthly ΔpCO_2 between September 2004 – July 2005 plotted as a function of SST at the K1 CELAS site

enhanced sea surface pCO_2 (high DIC) is expected following deep convective mixing and entrainment of subsurface CO_2 enriched water to the surface of the ocean. DIC supply to the surface is relatively low during summertime owing to biology and stratification of the ocean system. Additionally, sea-air ΔpCO_2 flux variability digressed towards negative flux following the outset of spring as the seawater surface starts to warm up. Thus, the ΔpCO_2 flux variability on an annual timescales generally indicates a consistent undersaturation of the NW subpolar site.

Seasonal sink estimates for the wintertime of 2004 and 2005 were calculated as 4.86 ± 0.15 and 4.15 ± 0.98 mol m^{-2} CO_2 a^{-1} respectively, while 0.29 ± 0.78 and 1.65 ± 1.40 mol m^{-2} a^{-1} were obtained for 2003 and 2004 summertime (Table 1). The CO_2 uptake of 2.36 ± 2.07 and 2.28 ± 1.34 mol m^{-2} CO_2 a^{-1} were computed for the autumn of 2003 and 2004 respectively. Based on the *in situ* observed data available, the short-term interseasonal CO_2 sink of the Porcupine Abyssal Plain time series site is estimated to have decreased by approximately 82.4, 3.6 and 17.2% for summer, autumn and wintertime respectively (Figure 4). Overall, there was persistent pCO_2 undersaturation of the surface seawater at the PAP site throughout the deployment periods, which

Table 1: Air–sea CO_2 flux estimates based on seasonal PAP data given in units of mol m^{-2} a^{-1}

Period	Air – sea CO_2 flux (mol m^{-2} a^{-1})*			
	Min.	Max.	Average	Std. Dev.
Summer 2003	−0.86	0.26	−0.29	0.78
Fall 2003	−4.35	−0.22	−2.36	2.07
Winter 2004	−5.00	−4.69	−4.86	0.15
Spring 2004	−4.58	−3.62	−4.03	0.49
Summer 2004	−3.22	−0.48	−1.65	1.40
Fall 2004	−3.23	−0.74	−2.28	1.34
Winter 2005	−4.77	−3.01	−4.15	0.98
Net CO_2 sink			−2.96	1.73

Note: *Negative indicates invasion of CO_2 into ocean from the atmosphere

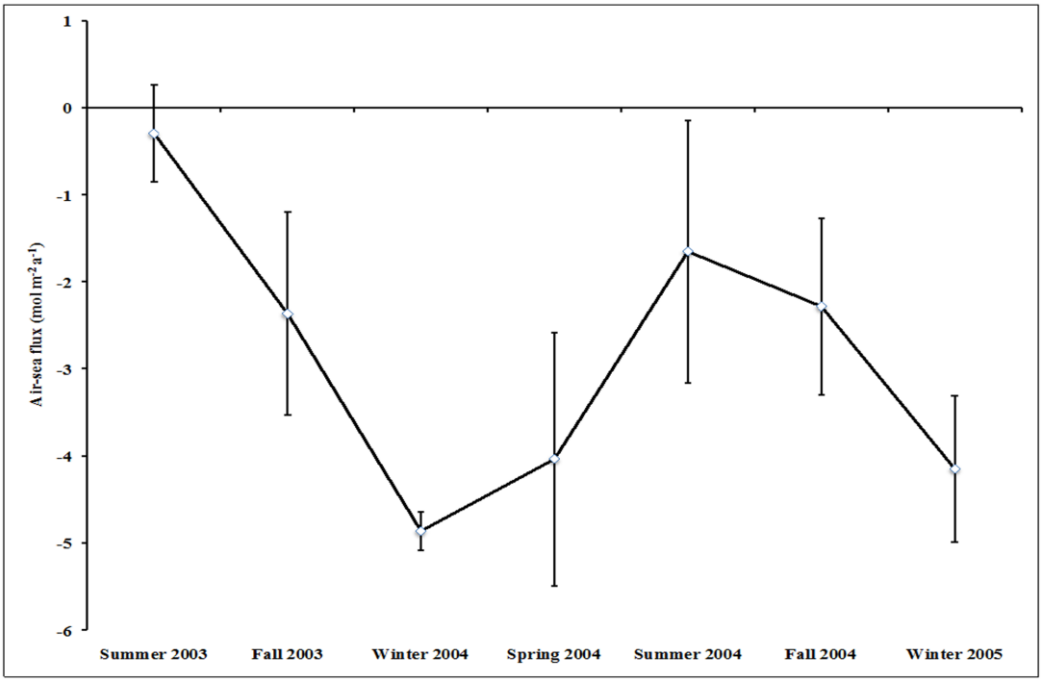

Figure 4 Interseasonal average air-sea CO_2 fluxes at PAP time series site (49 ^{o}N, 16.5 ^{o}W) between July 2003 and March 2005

approximates to annual CO_2 sink estimates of 2.06 ± 2.13, 3.21 ± 1.60 and 3.73 ± 0.84 mol m^{-2} CO_2 a^{-1} for the 2003, 2004 and 2005 deployments respectively. A net CO_2 uptake flux during the 2nd to 4th deployments is estimated to be -2.96 ± 1.73 mol m^{-2} CO_2 a^{-1}, indicating a perennial sink for the Northeast basin. However, a significant difference influx on an interseasonal timescale took place during the wintertime, which witnessed a net invasion (strong sink) of CO_2 compared to a moderate sink during the summertime.

The monthly air-sea CO_2 fluxes for the K1 CELAS observatory (56.5 ^{o}N, 52.6 ^{o}W) calculated based on observational data obtained between September 2004 and July 2005 is shown in Figure 5. The spatially monthly averaged air to sea fluxes indicate an oceanographic system that is perennially sequestering atmospheric CO_2 except in the peak of the wintertime (February/March 2005), when an estimated uptake of approximately 0.05 mol m^{-2} CO_2 a^{-1} invaded the seawater system, implying a relatively weak sink. During wintertime, surface water pCO_2 approached equilibrium with atmospheric CO_2. Overall, a net CO_2 uptake flux during the K1 CELAS SAMI deployments is estimated to be -1.84 ± 1.3 mol m^{-2} CO_2 a^{-1}, with a significantly strong sink capacity of 3.5 mol m^{-2} CO_2 a^{-1} obtained during late summer of 2004.

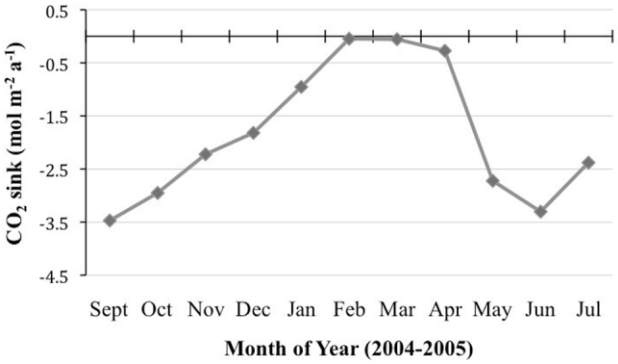

Figure 5 Monthly air-sea CO_2 fluxes at Northwest K1 CELAS time series site

However, the monthly air-sea CO_2 fluxes at the PAP time series site indicate a temperature dependence on the flux variability. This is an indication that the fluxes are controlled by *in situ* SST as suggested by the near-linear correspondence between monthly average SST and monthly average flux at the site (Figure 6). In the same way, to elucidate the mechanism that drives the monthly flux variability, which in turn influences the surface oceanic carbon cycle at the K1 CELAS site, a linear plot between CO_2 fluxes and SST is presented in Figure 7. The air-sea fluxes variability as shown by the non-dependence relationship between monthly average flux and SST reveals that the exchange is not entirely dominated by temperature- induced pCO_2-sw.

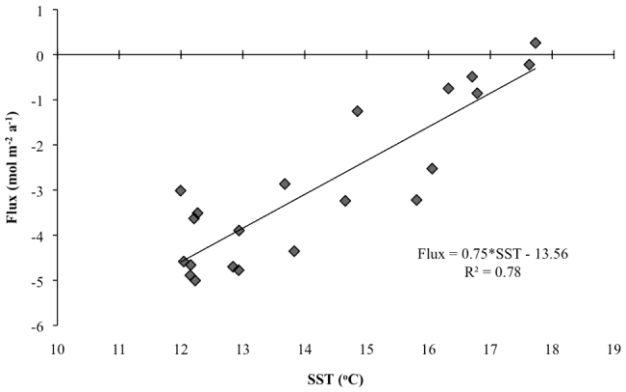

Figure 6 Monthly air-sea CO_2 fluxes versus SST at PAP site

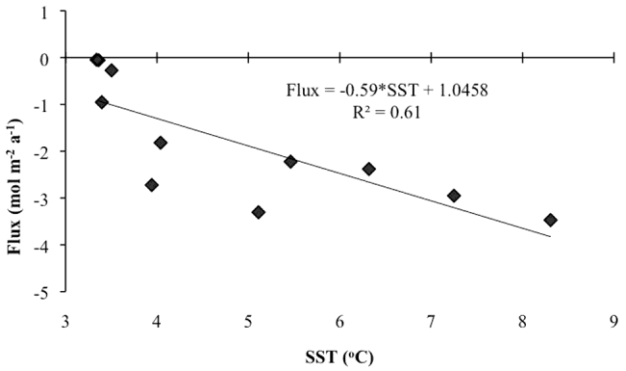

Figure 7 Monthly air-sea CO_2 fluxes versus SST at K1 CELAS site

3 Conclusions

This work indicates that the surface water pCO_2 cycle is characteristically marked by minimum and maximum pCO_2 levels for the summertime and wintertime respectively. There is a significant and consistent undersaturation of the PAP site of the North Atlantic Ocean. The pCO_2 concentration at the K1 CELAS demonstrated that the site is mostly undersaturated while exhibiting some degree of supersaturation between February and March 2005. Estimated net CO_2 uptake of 2.96 ± 1.73 and 1.84 ± 1.3 mol m^{-2} CO_2 a^{-1} were obtained during PAP (2nd - 4th) and K1 CELAS deployments respectively, thus indicating a regional perennial sink for CO_2. On an interseasonal timescale, significant difference in flux took place during the wintertime, which witnessed a strong pull of CO_2 compared to a moderate sink during the summertime. However, seasonal climatic changes in temperature, stratification, intense biological activities as well as convective mixing processes are identified as the primary drivers of air-sea flux variability for ocean carbon exchange in the region.

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