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Observed trends of $p\text{CO}_2$ and air-sea CO_2 fluxes in the North Atlantic Ocean

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Abstract Observed partial pressure of carbon dioxide ($p\text{CO}_2$) 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 $p\text{CO}_2$ of $335.9 \pm 89.6 \mu\text{atm}$ (2003), $286.7 \pm 103.5 \mu\text{atm}$ (2004), and $335.9 \pm 89.6 \mu\text{atm}$ (2005). The annual data for NW KI deployments indicated annual $p\text{CO}_2$ average of 336.6 ± 14.3 and $359.1 \pm 25.3 \mu\text{atm}$ for 2004 and 2005 respectively. The oceanic $p\text{CO}_2$ 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 $p\text{CO}_2$ 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 $1.84 \pm 1.3 \text{ mol m}^{-2} \text{ CO}_2 \text{ a}^{-1}$ were obtained for NE PAP (2nd - 4th) and NW KI deployments, respectively.

Keywords $p\text{CO}_2$; 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 $p\text{CO}_2$ existing between the atmosphere and surface of the ocean. Sea surface $p\text{CO}_2$ 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 $p\text{CO}_2$ 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 CO₂ 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 were obtained from the Porcupine Abyssal Plain and K1 Central Labrador Sea oceanographic mooring sites in the North Atlantic Ocean (Figure 1).

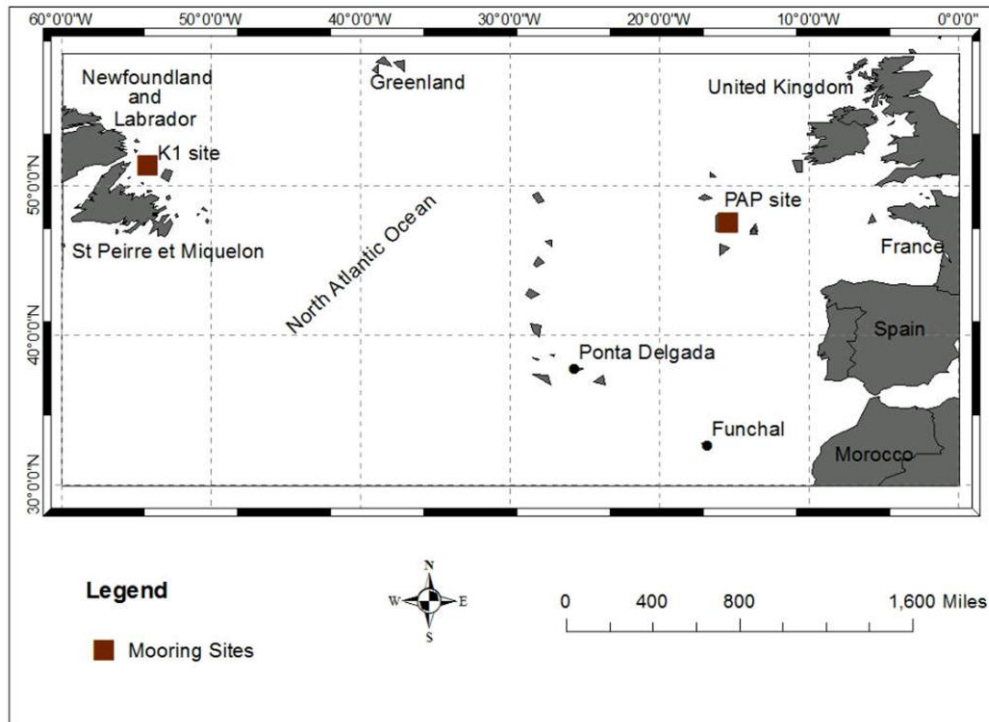


Figure 1 Map of the North Atlantic Ocean showing the Porcupine Abyssal Plain (49°N, 16.5°W) and Labrador Sea (56.5°N, 52.5°W)

1.2 Air-sea CO₂ fluxes estimation

The calculation of CO₂ flux (f_{CO_2}) in ocean and climate models is based on the indirect bulk method. The net exchange of CO₂ (f) between the ocean surface and the atmosphere is estimated from the air-sea difference in partial pressure of CO₂ (ΔpCO_2) and the gas transfer velocity (k) using the equation (Wanninkhof, 1992, 2007; Donelan and Wanninkhof, 2002):

$$f_{CO_2} = k \times (v) K_0 \times \Delta pCO_2 \quad (1)$$

Where k is the gas transfer velocity of CO₂ exchange, v is the wind speed, K_0 is the solubility of CO₂ in seawater and is a function of salinity and temperature (Weiss, 1974), and ΔpCO_2 is mean air-sea pCO_2 difference,

$$\Delta pCO_2 = [(pCO_{2-air}) - (pCO_{2-sw})] \quad (2)$$

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, v , 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 $Sc = 660$ (CO₂ at 20 C) have been proposed to describe k_{660} as a function of wind speed at a reference height of 10 m (v_{10}). Wanninkhof (1992) suggested a quadratic equation:

$$k_{660} = 0.31 v_{10}^2 \quad (3)$$

which gives k for any other Sc as

$$k = 0.31 v_{10}^2 \sqrt{660/Sc} \quad (4)$$

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

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

Moreover, recent investigation evaluating the transfer velocity-wind speed relationship using a long-term series of direct eddy correlation CO₂ flux measurements from the Baltic Sea suggests a combination of quadratic and linear wind speed dependence (Weiss et al., 2007; Rutgersson et al., 2008).

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

A positive flux (fCO_2) value represents a net CO₂ exchange from sea to the atmosphere and a negative flux value refers to the net CO₂ 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 CO₂ 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 CO₂ 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 CO₂-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.

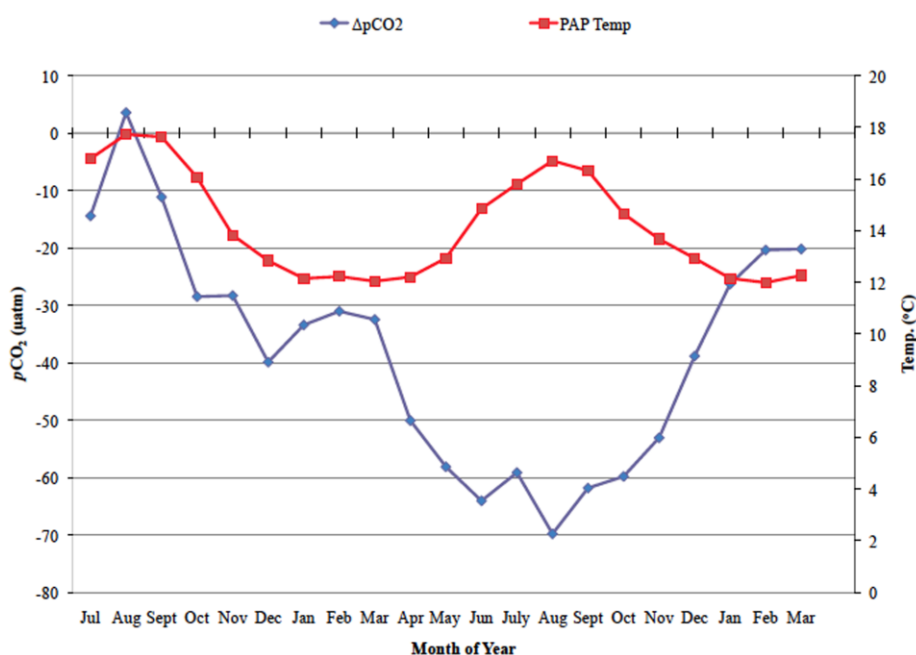


Figure 2 Monthly ΔpCO_2 between July 2003 – March 2005 plotted as a function of SST at the PAP observatory

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 CO₂ 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 CO₂ into the atmosphere leading to a decrease in net ocean CO₂ uptake during the summertime. This could be attributed to a dominant DIC-driven pCO_2 .

Considering the average monthly CO₂ 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 CO₂ flux (Figure 3). However, decreasing CO₂ sink of the time series location is observed following seasonal change from summertime of 2004 to wintertime. Uptake of CO₂ 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,

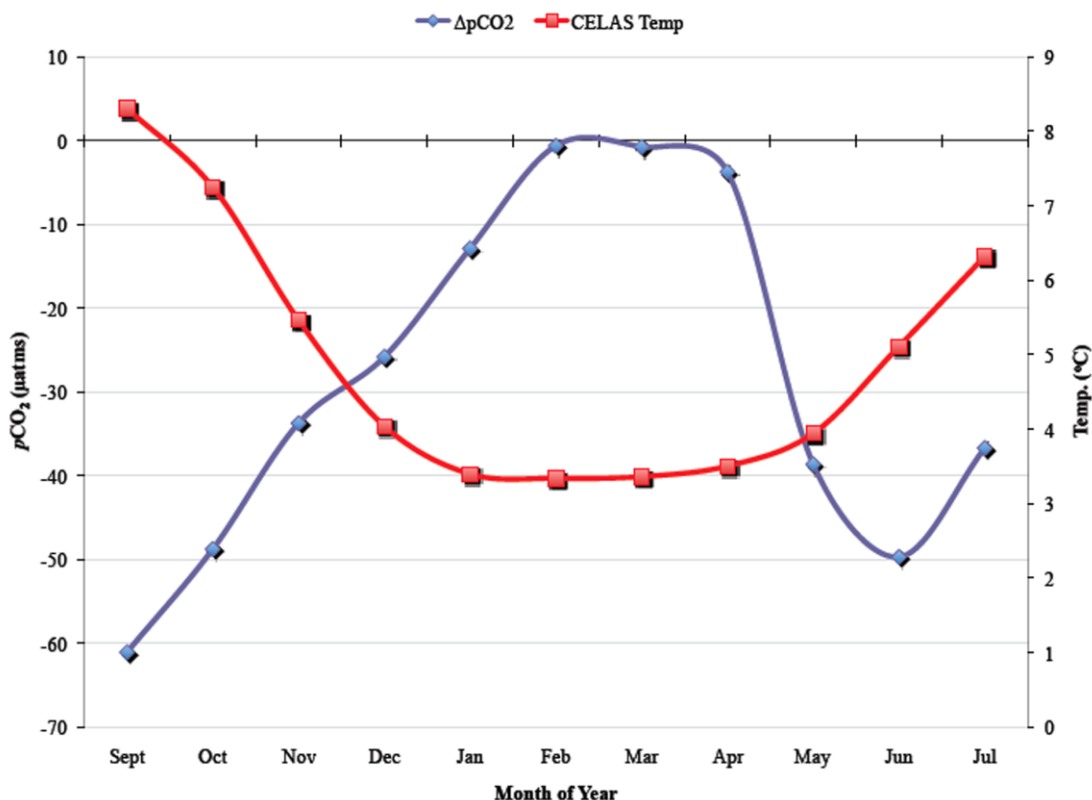


Figure 3 Monthly $\Delta p\text{CO}_2$ between September 2004 – July 2005 plotted as a function of SST at the K1 CELAS site

enhanced sea surface $p\text{CO}_2$ (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 $\Delta p\text{CO}_2$ flux variability digressed towards negative flux following the outset of spring as the seawater surface starts to warm up. Thus, the $\Delta p\text{CO}_2$ 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 $4.15 \pm$

$0.98 \text{ mol m}^{-2} \text{ CO}_2 \text{ a}^{-1}$ respectively, while 0.29 ± 0.78 and $1.65 \pm 1.40 \text{ mol m}^{-2} \text{ a}^{-1}$ were obtained for 2003 and 2004 summertime (Table 1). The CO_2 uptake of 2.36 ± 2.07 and $2.28 \pm 1.34 \text{ mol m}^{-2} \text{ CO}_2 \text{ a}^{-1}$ 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 $p\text{CO}_2$ 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 $\text{mol m}^{-2} \text{ a}^{-1}$

Period	Air – sea CO_2 flux ($\text{mol m}^{-2} \text{ a}^{-1}$)*			
	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

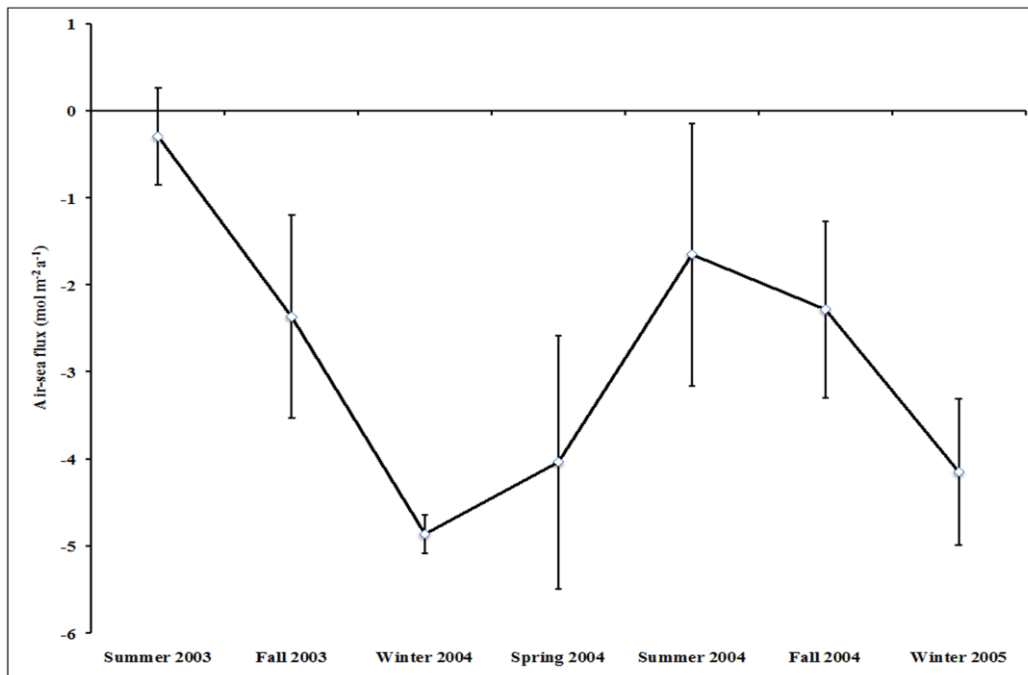


Figure 4 Interseasonal average air-sea CO₂ fluxes at PAP time series site (49 °N, 16.5 °W) between July 2003 and March 2005

approximates to annual CO₂ sink estimates of 2.06 ± 2.13 , 3.21 ± 1.60 and 3.73 ± 0.84 mol m⁻² CO₂ a⁻¹ for the 2003, 2004 and 2005 deployments respectively. A net CO₂ uptake flux during the 2nd to 4th deployments is estimated to be -2.96 ± 1.73 mol m⁻² CO₂ a⁻¹, indicating a perennial sink for the Northeast basin. However, a significant difference in flux on an interseasonal timescale took place during the wintertime, which witnessed a net invasion (strong sink) of CO₂ compared to a moderate sink during the summertime.

The monthly air-sea CO₂ fluxes for the K1 CELAS observatory (56.5 °N, 52.6 °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 CO₂ except in the peak of the wintertime (February/March 2005), when an estimated uptake of approximately 0.05 mol m⁻² CO₂ a⁻¹ invaded the seawater system, implying a relatively weak sink. During wintertime, surface water pCO₂ approached equilibrium with atmospheric CO₂. Overall, a net CO₂ uptake flux during the K1 CELAS SAMI deployments is estimated to be -1.84 ± 1.3 mol m⁻² CO₂ a⁻¹, with a significantly strong sink capacity of 3.5 mol m⁻² CO₂ a⁻¹ obtained during late summer of 2004.

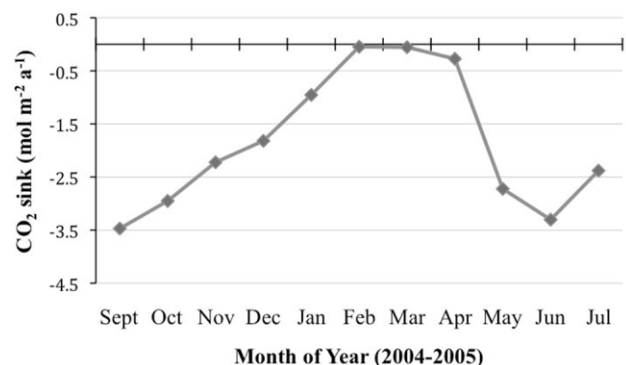


Figure 5 Monthly air-sea CO₂ fluxes at Northwest K1 CELAS time series site

However, the monthly air-sea CO₂ 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 CO₂ 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 pCO₂-sw.

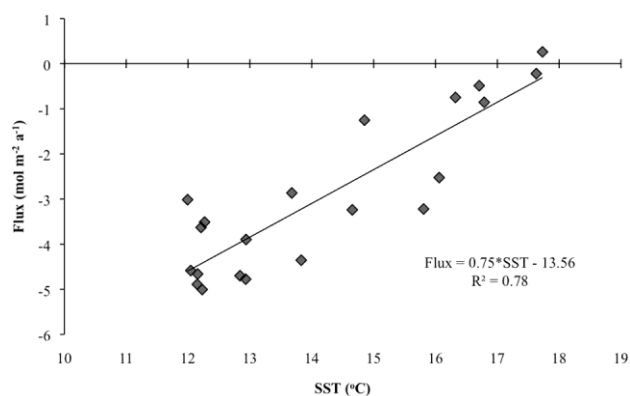


Figure 6 Monthly air-sea CO₂ fluxes versus SST at PAP site

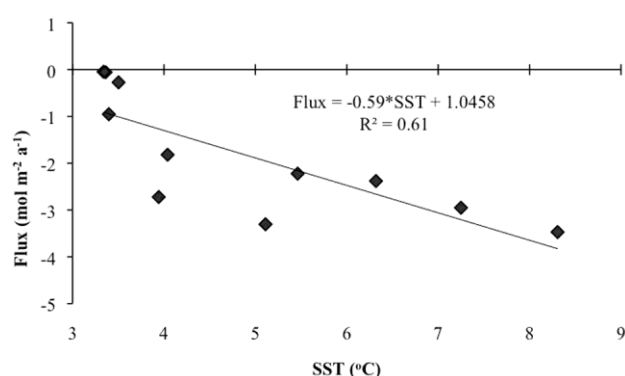


Figure 7 Monthly air-sea CO₂ fluxes versus SST at K1 CELAS site

3 Conclusions

This work indicates that the surface water $p\text{CO}_2$ cycle is characteristically marked by minimum and maximum $p\text{CO}_2$ levels for the summertime and wintertime respectively. There is a significant and consistent undersaturation of the PAP site of the North Atlantic Ocean. The $p\text{CO}_2$ 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 CO₂ uptake of 2.96 ± 1.73 and $1.84 \pm 1.3 \text{ mol m}^{-2} \text{ CO}_2 \text{ a}^{-1}$ were obtained during PAP (2nd - 4th) and K1 CELAS deployments respectively, thus indicating a regional perennial sink for CO₂. On an interseasonal timescale, significant difference in flux took place during the wintertime, which witnessed a strong pull of CO₂ 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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