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## Recent acceleration of the sea surface $f\text{CO}_2$ growth rate in the North Atlantic subpolar gyre (1993–2008) revealed by winter observations

Nicolas Metzl,<sup>1</sup> Antoine Corbière,<sup>1</sup> Gilles Reverdin,<sup>1</sup> Andrew Lenton,<sup>1,2</sup> Taro Takahashi,<sup>3</sup> Are Olsen,<sup>4,5</sup> Truls Johannessen,<sup>6</sup> Denis Pierrot,<sup>7</sup> Rik Wanninkhof,<sup>8</sup> Solveig R. Ólafsdóttir,<sup>9</sup> Jon Olafsson,<sup>9,10</sup> and Michel Ramonet<sup>11</sup>

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[1] Recent studies based on ocean and atmospheric carbon dioxide ( $\text{CO}_2$ ) observations, suggesting that the ocean carbon uptake has been reduced, may help explain the increase in the fraction of anthropogenic  $\text{CO}_2$  emissions that remain in the atmosphere. Is it a response to climate change or a signal of ocean natural variability or both? Regional process analyses are needed to follow the ocean carbon uptake and to enable better attributions of the observed changes. Here, we describe the evolution of the surface ocean  $\text{CO}_2$  fugacity ( $f\text{CO}_2^{\text{oc}}$ ) over the period 1993–2008 in the North Atlantic subpolar gyre (NASPG). This analysis is based primarily on observations of dissolved inorganic carbon (DIC) and total alkalinity (TA) conducted at different seasons in the NASPG between Iceland and Canada. The  $f\text{CO}_2^{\text{oc}}$  trends based on DIC and TA data are also compared with direct  $f\text{CO}_2$  measurements obtained between 2003 and 2007 in the same region. During winters 1993–2003, the  $f\text{CO}_2^{\text{oc}}$  growth rate was  $3.7 (\pm 0.6) \mu\text{atm yr}^{-1}$ , higher than in the atmosphere,  $1.8 (\pm 0.1) \mu\text{atm yr}^{-1}$ . This translates to a reduction of the ocean carbon uptake primarily explained by sea surface warming, up to  $0.24 (\pm 0.04) ^\circ\text{C yr}^{-1}$ . This warming is a consequence of advection of warm water northward from the North Atlantic into the Irminger basin, which occurred as the North Atlantic Oscillation (NAO) index moved into a negative phase in winter 1995/1996. In winter 2001–2008, the  $f\text{CO}_2^{\text{oc}}$  rise was particularly fast, between  $5.8 (\pm 1.1)$  and  $7.2 (\pm 1.3) \mu\text{atm yr}^{-1}$  depending on the region, more than twice the atmospheric growth rate of  $2.1 (\pm 0.2) \mu\text{atm yr}^{-1}$ , and in the winter of 2007–2008 the area was supersaturated with  $\text{CO}_2$ . As opposed to the 1990s, this appears to be almost entirely due to changes in seawater carbonate chemistry, the combination of increasing DIC and decreasing of TA. The rapid  $f\text{CO}_2^{\text{oc}}$  increase was not only driven by regional uptake of anthropogenic  $\text{CO}_2$  but was also likely controlled by a recent increase in convective processes—vertical mixing in the NASPG and cannot be directly associated with NAO variability. The  $f\text{CO}_2^{\text{oc}}$  increase observed in 2001–2008 leads to a significant drop in pH of  $-0.069 (\pm 0.007) \text{decade}^{-1}$ .

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### 1. Introduction

[2] Since the industrial revolution, the atmospheric concentration of carbon dioxide ( $\text{CO}_2$ ) has been rising from 280 ppm to its present concentration  $>386$  ppm in 2009, as a

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result of human activities (fossil fuel burning, cement production and land-use changes) [IPCC, 2007]. Approximately 50% of the anthropogenic  $\text{CO}_2$  released to the atmosphere has been captured and stored by the ocean [Sabine et al., 2004; Waugh et al., 2006] but it has been suggested that in recent decades the ocean carbon uptake has been reduced in the Southern Ocean [Le Quéré et al., 2007; Metzl, 2009], in the North Atlantic [Schuster et al., 2009 and references herein] and probably at global scale [Canadell et al., 2007; Le Quéré et al., 2009]. In the tropical Pacific, which has been intensively observed for more than two decades, trend analyses suggest that depending on the period and ocean circulation state, the ocean  $\text{CO}_2$  source increases or decreases [Feely et al., 2006; Ishii et al., 2009]. Understanding the historical decadal variations of the ocean carbon sink and how it will change in the future is important to better explain what drives climate and global biogeochemical changes, including the rate of ocean acidification and its potential impact on marine ecosystems. In this context, it is highly recommended to maintain long-term ocean  $\text{CO}_2$  observations (generally conducted at regional scale) to better understand what drives the variability of oceanic  $\text{CO}_2$  concentrations and associated air-sea  $\text{CO}_2$  fluxes [IOCCP, 2008; Doney et al., 2009] and synthesize these observations to reduce uncertainties of global carbon budget [Takahashi et al., 2009; Le Quéré et al., 2009].

[3] The North Atlantic Ocean (NA), north of  $50^\circ\text{N}$ , is considered as one of the stronger anthropogenic  $\text{CO}_2$  sinks [e.g., Sabine et al., 2004; Pérez et al., 2008; Vázquez-Rodríguez et al., 2009], a consequence of the large heat loss during winter and strong biological activity during spring and summer. The large-scale distribution of contemporary oceanic sinks and sources of atmospheric  $\text{CO}_2$  ( $\text{CO}_2^{\text{atm}}$ ) is now relatively well understood; it can be characterized by an uptake (sink) at high latitudes and outgassing (source) in the tropics [Takahashi et al., 2009]; globally the net oceanic uptake of anthropogenic  $\text{CO}_2$  is estimated to be  $-2.0 \pm 1.0 \text{ PgC yr}^{-1}$  ( $\text{Pg} = \text{Peta grams} = 10^{15} \text{ g}$ ). Observations indicate that the North Atlantic ( $>50^\circ\text{N}$ ) takes up  $0.28 \text{ PgC yr}^{-1}$  or  $\sim 15\%$  of the total annual oceanic uptake [Takahashi et al., 2009]. This region is also subject to large interannual variability, potentially related to large-scale atmospheric modes such as the North Atlantic Oscillation (NAO) and the Atlantic Multidecadal Variability (AMV) [Kushnir, 1994; Ting et al., 2009] governing basin-scale dynamical interactions between ocean circulation (advection and convection) and the atmosphere. The impact of these large-scale regimes on the mean annual air-sea  $\text{CO}_2$  fluxes and associated variabilities have not yet been clearly quantified and explained.

[4] Regional interannual and longer time scales studies in the NA indicate that the  $f\text{CO}_2$  of the sea surface has risen at a similar or faster rate than the atmospheric  $f\text{CO}_2$  over the last two to three decades [Bates, 2007; Corbière et al., 2007; Omar and Olsen, 2006; Olsen et al., 2006; Schuster and Watson, 2007; Schuster et al., 2009]. During the recent period, 1990–2006, this increase translates to a reduction of 50% of the total annual carbon uptake in the NA [Schuster et al., 2009]. This contradicts results from atmospheric inversion studies [Baker et al., 2006; Le Quéré et al., 2007;

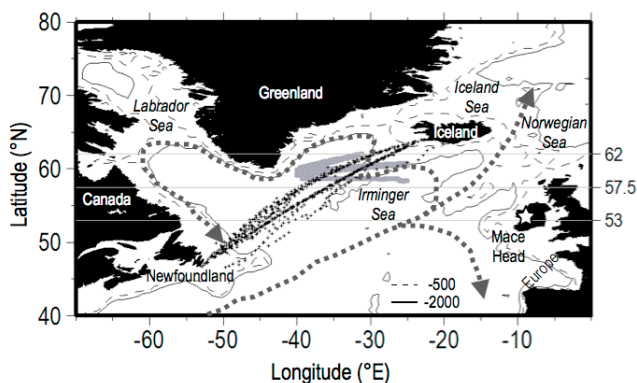
Rödenbeck, 2005] and ocean models [Raynaud et al., 2006; Thomas et al., 2008; Ullman et al., 2009; Wetzel et al., 2005] that suggest either significantly less variability or an increase in the strength of the NA ocean carbon sink. These differences could be explained by: i) the different periods considered; ii) different NAO and/or AMV phases; or iii) unidentified errors in both oceanic and atmospheric modeling experiments.

[5] A recent analysis based on 30 years of observations concluded that it was not possible to correlate  $f\text{CO}_2^{\text{oc}}$  variations with the NAO index directly due to limited sampling frequencies and large seasonal variability [Takahashi et al., 2009]. For this issue we have to ensure that appropriate observational systems are put in place to permit accurate quantification of the oceanic carbon sinks and detect changes reliably [Gruber, 2009; Watson et al., 2009]. In contrast, ocean biogeochemical models suggest that the NAO does affect  $f\text{CO}_2^{\text{oc}}$  and  $\text{CO}_2$  uptake in the Northern NA ( $>50^\circ\text{N}$ ), either through changes in: i) horizontal circulation and warming/cooling [Thomas et al., 2008]; or ii) vertical mixing and the supply of dissolved inorganic carbon (DIC) [Raynaud et al., 2006; Ullman et al., 2009]. In light of the significance of this region for the global carbon budget a more comprehensive understanding of the evolution of the integrated ocean carbon system, and its effect on the  $f\text{CO}_2^{\text{oc}}$  trend, is required.

[6] Our study aims to evaluate recent variations (1993–2008) of the surface  $\text{CO}_2$  system in the NA subpolar gyre (NASPG). In this respect it extends a previously published analysis based on DIC and Total Alkalinity (TA) observations obtained between 1993 and 2003 [Corbière et al., 2007]. As we are interested in investigating the long-term variations of DIC, TA and  $f\text{CO}_2^{\text{oc}}$ , we have selected data in the open ocean region ( $53\text{--}62^\circ\text{N}$  and  $45\text{--}25^\circ\text{W}$ ). Also, while data for all seasons will be shown, the trend analysis will be based on winter data only in order to avoid to potential biases due to the spatial heterogeneity that exist in the biological activity in the summer season. The decadal trends of observed oceanic carbon properties will be discussed in relation to their climatological and hydrographical context. Although ocean models are not perfect, recent simulations suggest that air-sea  $\text{CO}_2$  flux interannual anomalies are opposite in the northern and southern NASPG [Raynaud et al., 2006]. In our analysis, two periods, 1993–2003 and 2001–2008, and two regions are investigated (southern NASPG: sNASPG,  $53\text{--}57.5^\circ\text{N}$ ,  $45\text{--}35^\circ\text{W}$  and northern NASPG: nNASPG,  $57.5\text{--}62^\circ\text{N}$ ,  $40\text{--}25^\circ\text{W}$ ). The  $f\text{CO}_2^{\text{oc}}$  trends based on DIC and TA data will also be compared with direct  $f\text{CO}_2$  measurements obtained between 2003 and 2007 in the same regions. The decadal wintertime trends deduced from observed hydrological, nutrients and carbon data will then be used to separate the processes driving  $f\text{CO}_2^{\text{oc}}$  trends for each period. We will also compare  $f\text{CO}_2^{\text{oc}}$  trends deduced from independent data collected in the same region using different methods and in different periods.

## 2. Data and Methods

[7] Most of the data used in this work originate from SURATLANT (SURveillance de l'ATLANTique) cruises



**Figure 1.** The North Atlantic subpolar gyre showing the SURATLANT transect between Reykjavik (Iceland) and the Newfoundland (Canada) between 1993 and 2008. Each cross represents seawater surface sampling of carbon properties. Cruise tracks where  $f\text{CO}_2$  was measured continuously (NOAA/AOML) were superimposed on the SURATLANT transect. The gray thick lines correspond to the cruise of M/V *Nuka Arctica* where oceanic  $f\text{CO}_2$  were measured continuously in 2005 and 2007. The light gray lines show the open ocean region (53°N–62°N) and the southern (53°N–57.5°N) and northern (57.5°N–62°N) parts. The white star represents the Mace Head station in Ireland (53°20'N/9°54'W) where the atmospheric  $x\text{CO}_2$  was measured. Dashed lines indicate schematically main currents in the North Atlantic determined with altimetry data for 1992–2002 [Häkkinen and Rhines, 2004].

conducted in the NASPG between Reykjavik (Iceland) and Newfoundland (Canada) between 1993 and 2008 (Figure 1). These data were collected on instrumented merchant vessels crossing the subpolar gyre, each transit taking  $5 \pm 1$  days depending on weather conditions. During these cruises, usually every 3 months, SST (sea surface temperature) and SSS (sea surface salinity) were measured continuously, and DIC, TA and nutrients (nitrate, silicate and phosphate) samples were collected discretely (every 4 h) for subsequent analyses in land based laboratories. Since 2001, approximately 30 samples have been collected per cruise, and their DIC and TA were determined using a potentiometric method at the Laboratoire d'Océanographie et du Climat: Expérimentation et Approches Numériques (LOCEAN-IPSL, Paris, France) while nutrients (phosphate, nitrate, silicic acid) were analyzed at the Marine Research Institute (Reykjavik, Iceland). For DIC and TA, Certified Reference Materials provided by Prof. A.G. Dickson (Scripps Institution of Oceanography, San Diego, USA) were used for calibration. The uncertainty of the TA and DIC has been estimated to  $\pm 3 \mu\text{mol kg}^{-1}$ . Further details of the sampling methods and analytical techniques are described by Corbière *et al.* [2007]. Between 1993 and 1997, DIC was measured with a coulometric method [Chipman *et al.*, 1993], while TA was reconstructed from a TA-SSS relationship [Corbière *et al.*, 2007]. The  $f\text{CO}_2^{\text{oc}}$  values were computed from DIC-TA pairs and phosphate and silicate concentrations (observed or climatological values) at in situ

SST and SSS; for these calculations we used the CO2SYS program [Pierrot *et al.*, 2006]. The  $\text{CO}_2$  solubility coefficients were taken from Weiss [1974] and the dissociation constants from Mehrbach *et al.* [1973] refitted by Dickson and Millero [1987], following the recommendations of Wanninkhof *et al.* [1999] and Lee *et al.* [2000]. Taking into account DIC and TA uncertainties, following Omar *et al.* [2003], the overall accuracy of calculated  $f\text{CO}_2$  is estimated to be 7–8  $\mu\text{atm}$ .

[8] The calculated  $f\text{CO}_2$  was compared with direct oceanic  $f\text{CO}_2$  measurements collected on-board the M/V *Skògafoss* and M/V *Nuka Arctica* (Figure 1). The *Skògafoss* operates along the SURATLANT transects between Iceland and Newfoundland and collects data as part of the *CO2 on ships* program funded by NOAA, whereas *Nuka* crosses the Atlantic between Denmark and Greenland along approximately 60°N [Olsen *et al.*, 2008] (Figure 1) and collects data as part of the EU IP CARBOOCEAN. The systems on each of the ships are similar, measuring the  $\text{CO}_2$  concentration in a headspace in equilibrium with a continuous stream of seawater and is calibrated with reference gases traceable to the WMO scale, see Pierrot *et al.* [2009] for a full description of this method. The uncertainty of the  $f\text{CO}_2$  measurements for these systems is estimated to be  $\pm 1 \mu\text{atm}$ , however based on intercomparison analysis, including errors in equilibrator and in situ temperature records, the absolute uncertainty of  $f\text{CO}_2$  is  $\pm 4 \mu\text{atm}$  [IOCCP, 2008]. For the *Skogafoss* we use the data collected between 2003 and 2007, and for *Nuka* we use the data collected in 2005 and 2007, no data were obtained in this area during the winter of 2006 as none of the ships operated on this route at that time.

[9] The differences between measured and calculated oceanic  $f\text{CO}_2$  have been evaluated using collocated samples collected on several SURATLANT cruises (February, May, July and November;  $n = 54$ ). The mean difference ( $f\text{CO}_{2\text{mes}} - f\text{CO}_{2\text{calc}}$ ) was calculated to be 2.3 ( $\pm 11.1$ )  $\mu\text{atm}$  which is slightly higher than the 1.8 ( $\pm 8.3$ )  $\mu\text{atm}$  determined by Wanninkhof *et al.* [1999] in the North Atlantic. This difference and larger uncertainty may reflect the larger sample size ( $n = 99$ ) used by Wanninkhof *et al.* [1999] than our study which compared several cruises, different ships and periods. It is important to note that the differences do not exhibit any systematic trend over the study period, and this adds confidence to the evaluation of interannual variability.

[10] To compare the trends of oceanic and atmospheric  $f\text{CO}_2$  we used atmospheric  $\text{CO}_2$  concentrations continuously measured since 1992 at the Mace Head station in Ireland (53°20'N–9°54'W) and converted monthly mean of  $x\text{CO}_2^{\text{atm}}$  to  $f\text{CO}_2^{\text{atm}}$  as recommended by Dickson *et al.* [2007].

### 3. Results

#### 3.1. Data Selection

[11] The NASPG is a site of mixing of surface waters originating from the subtropical, subpolar and polar regions. The distribution and evolution of its sea surface properties are strongly influenced by two currents: i) the North Atlantic Current (NAC), becoming the Irminger Current, carry warm and salty water around its southern and eastern rims; and ii)

the Arctic Ocean surface outflow from the Greenland and Labrador seas delivering cold and low-salinity water (this circulation is principally concentrated on the continental shelves and slopes of East Greenland and Newfoundland) along the western side of the gyre. We divided the area covered by the SURATLANT transect into two regions corresponding to increasing distance from the warm and salty Irminger Current: the northern NASPG (nNASPG, 57.5–62°N), and southern NASPG (sNASPG, 53–57.5°N).

[12] In a first approach, a direct estimate of  $f\text{CO}_2^{\text{oc}}$  regional trends using all winter cruises 1993–2008 leads to an increase of  $+4.3 (\pm 0.4) \mu\text{atm yr}^{-1}$  in the nNASPG and  $4.7 (\pm 0.5) \mu\text{atm yr}^{-1}$  sNASPG suggesting that i) the  $f\text{CO}_2^{\text{oc}}$  increase was faster than in the atmosphere and ii) the  $f\text{CO}_2^{\text{oc}}$  rate was slightly higher in the southern region. This first estimate suggests that the  $f\text{CO}_2^{\text{oc}}$  increasing rate over 1993–2008 is high compared to results obtained for the whole domain (nNASPG + sNASPG) and for different periods, e.g., 1972–1990 [Omar and Olsen, 2006] or 1993–2003 [Corbière et al., 2007]. Therefore the persistence and characteristics of the decadal variability will be tested by investigating trends over two periods 1993–2003 and 2001–2008. All data are presented for each region in Figures 2 and 3, and winter trends for the two periods are listed in Table 1. Figure 3 also includes winter values of the NAO index that was more variable in 1993–2001 than after 2001. Finally, as indicated in the method section, only in 2001–2008 we were able to measure SST, SSS, DIC, TA and nutrients properties, whereas at the start of the SURATLANT experiment only SST, SSS and DIC were regularly measured. This is also why we prefer to investigate the surface ocean properties trends over two distinct periods that also permit to compare trends deduced from calculated and measured  $f\text{CO}_2^{\text{oc}}$ .

### 3.2. Period 1993–2003

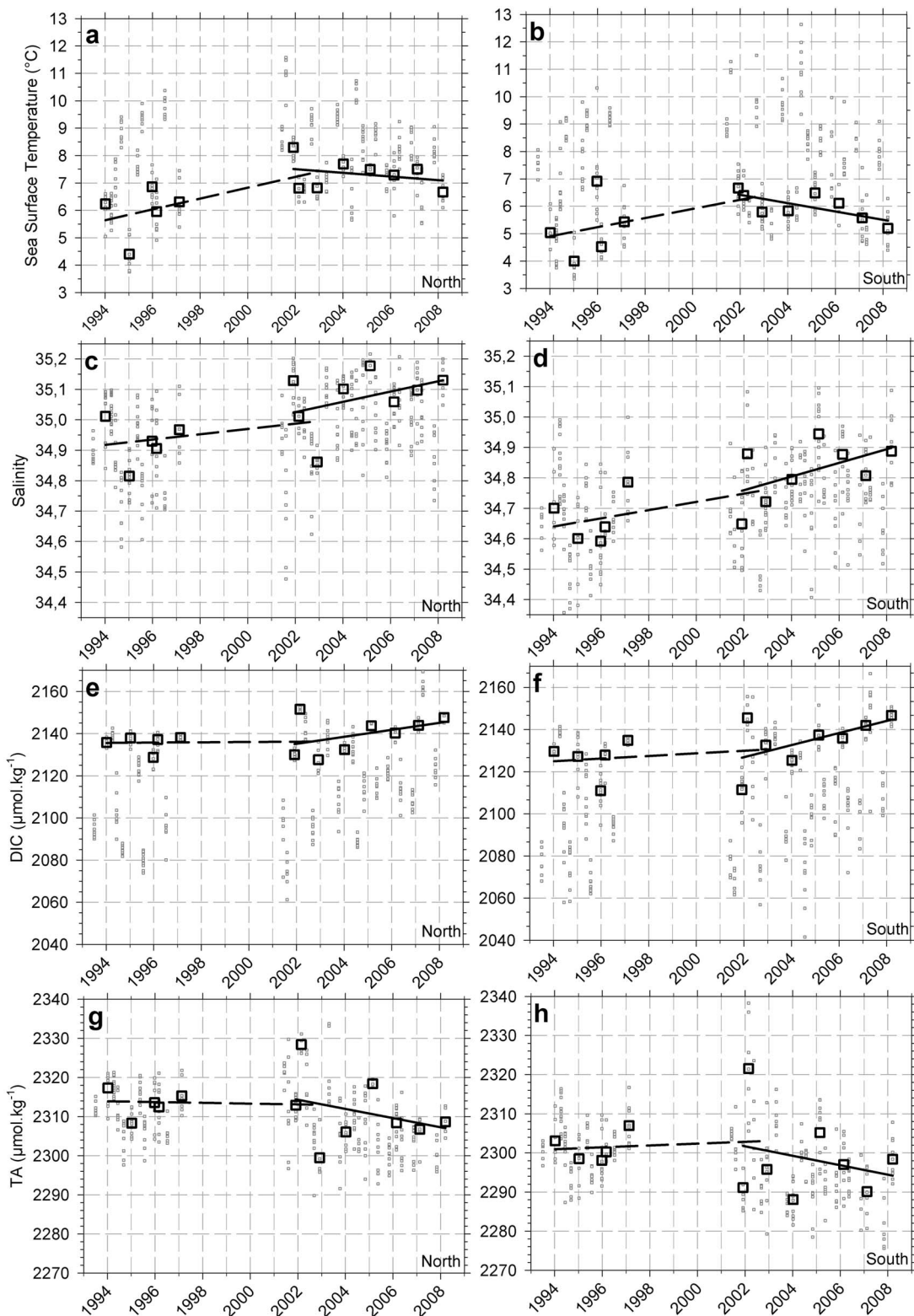
[13] The surface layer of the NASPG became warmer and more saline during this period. Both regions (nNASPG and sNASPG) exhibit increasing winter SST and SSS (Table 1) with large interannual variations (Figure 2). This net increase is the result of circulation changes associated with the shift from a high positive NAO index in the early 1990s to a negative index in winter 1995/1996 (Figures 3a and 3c). The period 1995/1996 was also marked by a change in multidecadal Atlantic variability mode (AMV) from negative (1966–1996) to positive index (1996–2008) [Ting et al., 2009; Reverdin, 2010]. This change in the AMV caused initially an anomalously strong northward transport of the North Atlantic Current followed by a weaker circulation and a contraction of the NASPG [Häkkinen et al., 2008]. During the period of negative (or neutral) NAO index a weak dynamic height gradient acts to contract the subpolar gyre and more subtropical water is carried into the northeastern region of the subpolar gyre [Häkkinen and Rhines, 2004; Hátún et al., 2005]. However, it has been also observed and simulated that the gyre circulation does not follow the NAO pattern very directly [Häkkinen and Rhines, 2004; Herbaut and Houssais, 2009]. Consequently, the air-sea  $\text{CO}_2$  flux - NAO correlation appears to be strongest with 2 to 4 years phase lag [Thomas et al., 2008]. Observations of Subpolar Mode Water (SMW) made between 1996 and 2003 in this

region [Thierry et al., 2008] also reveal a significant warming of  $1.41^\circ\text{C}$  ( $0.2^\circ\text{C yr}^{-1}$ ) and salinity increase of  $0.11$  ( $0.02 \text{ yr}^{-1}$ ) that is close to what we observe in surface water during winters of 1993–2003 (Table 1). After a second NAO shift in 2000/2001 (from positive to negative), the warm anomalies persisted in the NASPG in 2001–2003. In the nNASPG, the warming observed in our data ( $0.24 \pm 0.04^\circ\text{C yr}^{-1}$ ) is coherent with that observed further east along the eastern flank of the Reykjanes Ridge [Thierry et al., 2008], where the North Atlantic Current (NAC) flows into the Irminger and the Nordic Seas. Although the sNASPG is less affected by this water mass advection than nNASPG, the changes are fairly similar, suggesting that the circulation changes and positive trends of SST and SSS impact  $f\text{CO}_2$  over the entire NASPG.

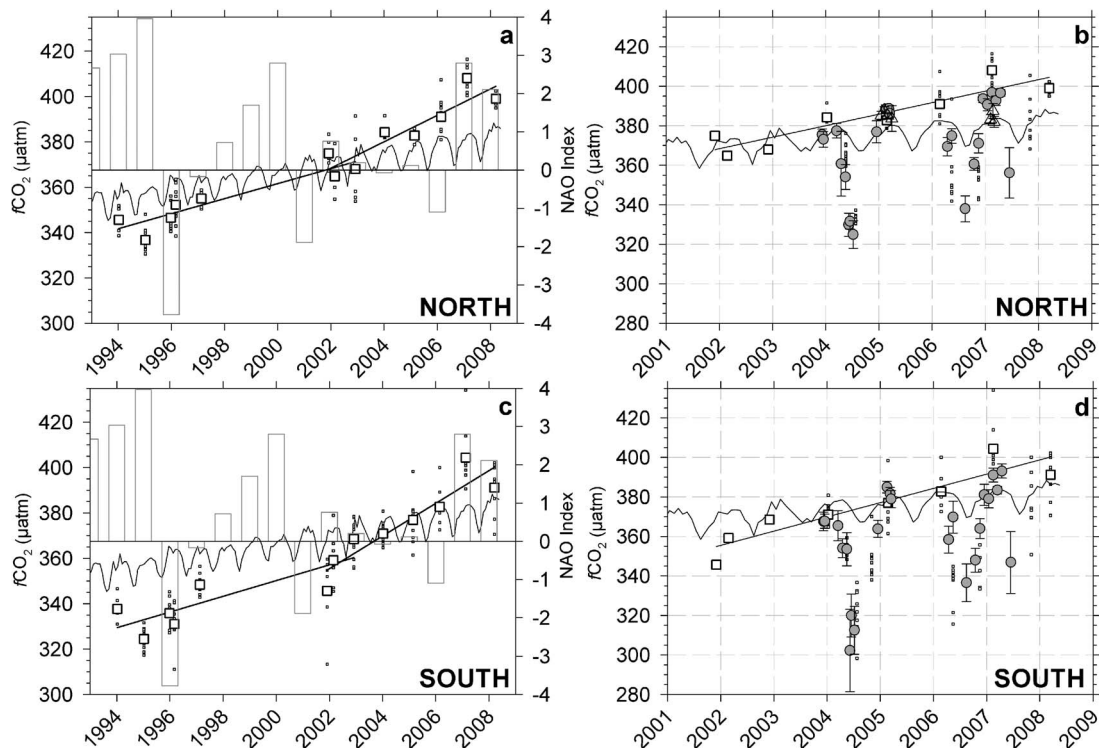
[14] During 1993–2003 the winter DIC data displays almost no trend (Figure 2 and Table 1): a small annual DIC decrease of  $-0.29 \pm 0.26 \mu\text{mol kg}^{-1}$  in the nNASPG (not statistically significant) and a small DIC increase of  $0.66 \pm 0.45 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  in the sNASPG. Corbière et al. [2007] suggested this may be the result of the increase in DIC due to  $\text{CO}_2$  uptake (between  $1$  and  $2 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$ ) counterbalanced by an increase in advection of low-DIC NAC waters into the NASPG. This compensation of DIC increase due to anthropogenic  $\text{CO}_2$  invasion by horizontal advection was also identified in interannual biogeochemical simulations of the North Atlantic [Thomas et al., 2008]. An alternative explanation is a decrease in vertical mixing, after 1995 [Lazier et al., 2002; Thierry et al., 2008; Ullman et al., 2009], which would also result in lower surface DIC (as less DIC would be imported from deep layers).

[15] Although TA was not measured at the start of the SURATLANT time series (1993–1997), we can predict TA distribution based on TA/SSS relationship from seasonal observations in 2001–2002 [Corbière et al., 2007]. For salinity between 34.5 and 35.3, the TA/SSS relationship ( $\text{TA} = 45.808 \times \text{SSS} + 713.5$ ,  $r^2 = 0.92$ ;  $\text{RMSD} = \pm 10.3 \mu\text{mol kg}^{-1}$ ), was found to be close to the one derived for the entire Atlantic Ocean [Millero et al., 1998]. Over 1993–1997, TA is thus directly estimated from SSS which increases, whereas in 2001–2003 we used winter TA measurements to evaluate the trends. This leads to almost no trend in TA for winters 1993–2003 in both regions (Table 1 and Figure 2), and hence has very little impact on  $f\text{CO}_2^{\text{oc}}$  trend for 1993–2003.

[16] Corbière et al. [2007] showed that winter  $f\text{CO}_2^{\text{oc}}$ , averaged over the NASPG open ocean area, increased at a rate of  $2.8 \pm 0.9 \mu\text{atm yr}^{-1}$ , based on calculated  $f\text{CO}_2^{\text{oc}}$  but also reconstructed winter  $f\text{CO}_2^{\text{oc}}$  for the period 1997–2000 with no DIC data. In the current study, we have calculated the  $f\text{CO}_2^{\text{oc}}$  trends in the nNASPG and sNASPG using only observed DIC/TA pairs and TA reconstructed for 1993–1997 (Table 1 and Figures 3a and 3c). The  $f\text{CO}_2^{\text{oc}}$  trend for winter is now estimated to be around  $3.7 (\pm 0.6) \mu\text{atm yr}^{-1}$  over 1993 to 2003 in both regions (i.e., two times higher than in the atmosphere;  $1.8 \mu\text{atm yr}^{-1}$ ) and close to the  $f\text{CO}_2^{\text{oc}}$  growth rate previously determined ( $+2.8 \mu\text{atm yr}^{-1}$ ) [Corbière et al., 2007]. To investigate the error of the  $f\text{CO}_2^{\text{oc}}$  trend due to missing TA observations in 1993–1997, we also estimate the trend using the reconstructed TA based on the relationship proposed by Lee et al. [2006] that includes



**Figure 2.** Interannual variability of Sea Surface Temperature (SST), Sea Surface Salinity (SSS), Dissolved Inorganic carbon (DIC) and Total Alkalinity (TA) between 1993 and 2008. The in situ observations are indicated with small gray squares. Only the data from December through March were used to calculate winter trends (black dashed line for the period 1993–2003 while the solid black line is for the 2001–2008). The black open squares indicate the monthly mean values for each cruise in the (a, c, e, and g) northern and (b, d, f, and h) southern regions.



**Figure 3.** Winter trends of sea surface  $f\text{CO}_2$  for 1993–2003 and 2001–2008 (black lines) in the north Atlantic subpolar gyre (NASPG). The winter (December to March) in situ observations used for estimating the linear trends are indicated (dots). Open squares indicate ocean  $f\text{CO}_2$  averaged for each cruise between (a, b)  $57.5^\circ\text{N}$ – $62^\circ\text{N}$ , nNASPG and between (c, d)  $53^\circ\text{N}$ – $57.5^\circ\text{N}$ , sNASPG. The atmospheric  $f\text{CO}_2$  deduced from concentrations observed at the Mace Head station in Ireland is also represented (thin line). In Figures 3a and 3c, the bars represent the NAO index (<http://www.cgd.ucar.edu/cas/jhurrell/>). In Figures 3b and 3d, gray dots indicate for the same region the oceanic  $f\text{CO}_2$  measured continuously onboard the *M/V Skogafoss* (AOML/NOAA), and the triangles represent the oceanic  $f\text{CO}_2$  measured continuously onboard the *M/V Nuka Arctica*. Vertical bars indicate the standard deviation around the mean.

**Table 1.** Winter Trends (1993–2003 and 2001–2008) of Sea Surface Temperature, Salinity, DIC, Normalized DIC (nDIC), TA, Normalized TA (nTA), Atmospheric  $f\text{CO}_2$  ( $f\text{CO}_2^{\text{atm}}$ ), Oceanic  $f\text{CO}_2$  Calculated ( $f\text{CO}_2^{\text{calc}}$ ) and Measured ( $f\text{CO}_2^{\text{meas}}$ ) in the North Atlantic Subpolar Gyre

	1993–2003		2001–2008	
	North $57.5$ – $62^\circ\text{N}$	South $53$ – $57.5^\circ\text{N}$	North $57.5$ – $62^\circ\text{N}$	South $53$ – $57.5^\circ\text{N}$
Temperature ( $^\circ\text{C yr}^{-1}$ )	$0.24 \pm 0.04$	$0.14 \pm 0.04$	$-0.10 \pm 0.04$	$-0.14 \pm 0.04$
Salinity ( $\text{yr}^{-1}$ )	$0.013 \pm 0.005$	$0.013 \pm 0.005$	$0.010 \pm 0.006$	$0.026 \pm 0.007$
DIC ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ )	$-0.29 \pm 0.26$	$0.66 \pm 0.45$	$2.15 \pm 0.40$	$3.19 \pm 0.53$
nDIC ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ )	$-1.09 \pm 0.39$	$-0.12 \pm 0.34$	$1.55 \pm 0.52$	$1.65 \pm 0.45$
TA ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ )	$-0.17 \pm 0.30^{\text{a}}$	$0.02 \pm 0.37^{\text{a}}$	$-0.95 \pm 0.44$	$-0.73 \pm 0.62$
nTA ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ )	$-1.04 \pm 0.24$	$-0.57 \pm 0.41$	$-1.59 \pm 0.42$	$-2.37 \pm 0.59$
Phosph ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ )	-	-	$0.028 \pm 0.004$	$0.036 \pm 0.005$
Nitr ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ )	-	-	$0.35 \pm 0.07$	$0.58 \pm 0.09$
Sil ( $\mu\text{mol kg}^{-1} \text{yr}^{-1}$ )	-	-	$0.26 \pm 0.05$	$0.40 \pm 0.05$
$f\text{CO}_2^{\text{atm}}$ ( $\mu\text{atm yr}^{-1}$ )		$1.8 \pm 0.1^{\text{b}}$		$2.1 \pm 0.2^{\text{b}}$
$f\text{CO}_2^{\text{calc}}$ ( $\mu\text{atm yr}^{-1}$ )	$3.7 \pm 0.5$	$3.8 \pm 0.7$	$5.8 \pm 1.1$	$7.2 \pm 1.3$
			$7.1 \pm 1.1^{\text{c}}$	$5.5 \pm 1.6^{\text{c}}$
$f\text{CO}_2^{\text{meas}}$ ( $\mu\text{atm yr}^{-1}$ )	-	-	$5.7 \pm 0.9^{\text{d}}$	$4.8 \pm 1.4^{\text{d}}$

<sup>a</sup>TA was simulated over the period 1993–1997 (see text and Corbière et al. [2007]).

<sup>b</sup>Mace Head Station ( $53^\circ 20'\text{N}/9^\circ 54'\text{W}$ , Ireland).

<sup>c</sup> $f\text{CO}_2^{\text{calc}}$  calculated from January 2004 to February 2007.

<sup>d</sup> $f\text{CO}_2^{\text{meas}}$  measured from December 2003 to March 2007.

constraints on both SSS and SST. This results in lower estimated TA than from the TA–SSS relationship of *Corbière et al.* [2007], and hence derived  $f\text{CO}_2^{\text{oc}}$  is higher for the period 1993–1997. Consequently, the  $f\text{CO}_2^{\text{oc}}$  trend is smaller than our first estimate of  $3.7 \mu\text{atm yr}^{-1}$  (i.e.,  $2.6 (\pm 0.4) \mu\text{atm yr}^{-1}$  in the sNASPG and  $2.1 (\pm 0.3) \mu\text{atm yr}^{-1}$  in the nNASPG), but importantly still above the atmospheric  $\text{CO}_2$  increase. Although the methods of calculation and data selection are different, the present results confirm previous analyses in the NASPG [*Corbière et al.*, 2007; *Schuster et al.*, 2009], i.e., a decreasing oceanic  $\text{CO}_2$  sink during the nineties, when the winter NAO index decreased from high positive index around 1989–1995 to neutral or small positive/negative values in mid-nineties. We will now explore if this trend has persisted in recent years when the NAO index variations were not as large as observed (in the mid-nineties), but still corresponds to a positive AMV index [e.g., *Ting et al.*, 2009; *Reverdin*, 2010].

### 3.3. Period 2001–2008

[17] After the second NAO shift (from positive to negative) in winter 2000/2001, the NAO index remained close to 0 (until winter 2005/2006; Figures 3a and 3c). In the period 2001–2008, the surface waters were clearly warmer than in 1993–1997 but appear to be becoming cooler (about  $-0.1^\circ\text{C yr}^{-1}$ ; Table 1 and Figure 2). Conversely, the salinity rose at about the same rate as in 1993–2003 in the nNASPG ( $0.01 \text{ yr}^{-1}$ ) but faster in the sNASPG ( $0.026 \text{ yr}^{-1}$ ). Winter data also indicate an increase of all nutrients in both regions (Table 1) and consistent with salinity, the increasing concentrations of nutrients appear to have been faster in the sNASPG than the nNASPG.

[18] In contrast to 1993–2003, both DIC and TA changed over the 2001–2008 period; DIC increased whereas TA decreased. The trends in DIC were  $2.15 (\pm 0.40) \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  in the nNASPG and  $3.19 (\pm 0.53) \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  in the sNASPG which is larger than the DIC increase expected from anthropogenic  $\text{CO}_2$  ocean uptake in this region [*Friis et al.*, 2005; *Olsen et al.*, 2006; *Pérez et al.*, 2008; *Vázquez-Rodríguez et al.*, 2009]. To remove the variations due to the salinity increase we evaluated the trends for salinity normalized DIC (nDIC) at  $1.6 (\pm 0.5) \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  in the NASPG. The magnitude of this trend is very close to results discussed by *Skjelvan et al.* [2008] who reported an nDIC increase of  $1.3 (\pm 0.7) \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  in the surface waters of the Norwegian Sea (2001–2006 observations at Ocean Weather Station M,  $66^\circ\text{N}/2^\circ\text{E}$ ). In the nNASPG, the observed decrease in TA for years 2001–2008 reaches almost  $-1 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  and is opposite in sign to that observed in SSS; consequently the salinity normalized TA (nTA) decreases at a faster rate than TA (up to  $-2.4 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$  in the sNASPG).

[19] The increase in DIC and decrease in TA have a combined effect, increasing  $f\text{CO}_2^{\text{oc}}$ . Consequently the  $f\text{CO}_2^{\text{oc}}$  trends estimated for the period 2001–2008 indicate a very rapid evolution, approximately 3 times faster than in the atmosphere (Table 1 and Figure 3). The  $f\text{CO}_2^{\text{oc}}$  growth rate is higher in the sNASPG ( $+7.2 \pm 1.3 \mu\text{atm yr}^{-1}$ ) than in the nNASPG ( $+5.8 \pm 1.1 \mu\text{atm yr}^{-1}$ ) which is mainly explained by the difference of the DIC trends in these two regions.

These results, based on calculated  $f\text{CO}_2^{\text{oc}}$ , are confirmed by comparison with concomitant oceanic  $f\text{CO}_2$  measurements made since the end of the year 2003 (Table 1 and Figures 3b and 3d). The linear trends for calculated  $f\text{CO}_2^{\text{oc}}$  are statistically indistinguishable from those obtained from measured  $f\text{CO}_2^{\text{oc}}$  for the period from December 2003 through February 2007 (Table 1). The rapid  $f\text{CO}_2^{\text{oc}}$  growth over 7 years represents an increase of  $\sim 45 \mu\text{atm}$ . Over the period 2007–2008,  $f\text{CO}_2^{\text{oc}}$  was on average between 390 and 410  $\mu\text{atm}$ , with local values up to 434  $\mu\text{atm}$  in February 2007, i.e., well above the equilibrium ( $\Delta f\text{CO}_2 \sim +50 \mu\text{atm}$ ). High  $f\text{CO}_2^{\text{oc}}$  (above 430  $\mu\text{atm}$ ) of this magnitude has never been observed in the NASPG and correspond to the NASPG acting a oceanic source of atmospheric  $\text{CO}_2$ .

## 4. Discussion: Drivers of the Surface $f\text{CO}_2^{\text{oc}}$ Evolution

[20] The  $f\text{CO}_2^{\text{oc}}$  trends estimated for winter in the nNASPG and sNASPG were clearly higher in 2001–2008 when compared to 1993–2003 (Figures 3a and 3c and Table 1). To explore the drivers of this recent  $f\text{CO}_2^{\text{oc}}$  acceleration we have estimated the changes in  $f\text{CO}_2^{\text{oc}}$  contributed by four major quantities: SST, SSS, DIC and TA using the following equation:

$$\begin{aligned} d f\text{CO}_2/d t &= (\partial f\text{CO}_2/\partial \text{SST})(d\text{SST}/dt) + (\partial f\text{CO}_2/\partial \text{SSS})(d\text{SSS}/dt) \\ &+ (\partial f\text{CO}_2/\partial \text{DIC})(d\text{DIC}/dt) + (\partial f\text{CO}_2/\partial \text{TA})(d\text{TA}/dt) \end{aligned} \quad (1)$$

[21] This equation is rearranged as a function of known quantities:

$$\begin{aligned} d f\text{CO}_2/d t &= \tau(\langle f\text{CO}_2 \rangle)(d\text{SST}/dt) + \eta(\langle f\text{CO}_2 \rangle/\langle \text{SSS} \rangle)(d\text{SSS}/dt) \\ &+ \gamma(\langle f\text{CO}_2 \rangle/\langle \text{DIC} \rangle)(d\text{DIC}/dt) + \Gamma(\langle f\text{CO}_2 \rangle/\langle \text{TA} \rangle)(d\text{TA}/dt) \end{aligned} \quad (2)$$

where  $\langle X \rangle$  represents the mean winter values for nNASPG or sNASPG;

$t$  is time

$\tau = (\partial \ln f\text{CO}_2/\partial \text{SST}) = (\partial f\text{CO}_2/\langle f\text{CO}_2 \rangle)/\partial \text{SST}$ ;  $\tau = 0.0423$  per  $^\circ\text{C}$  [*Takahashi et al.*, 1993]

$\eta = (d \ln f\text{CO}_2/d \ln \text{SSS}) = (\partial f\text{CO}_2/\partial \text{SSS}) (\langle \text{SSS} \rangle/\langle f\text{CO}_2 \rangle)$ ;  $\eta = 0.9$

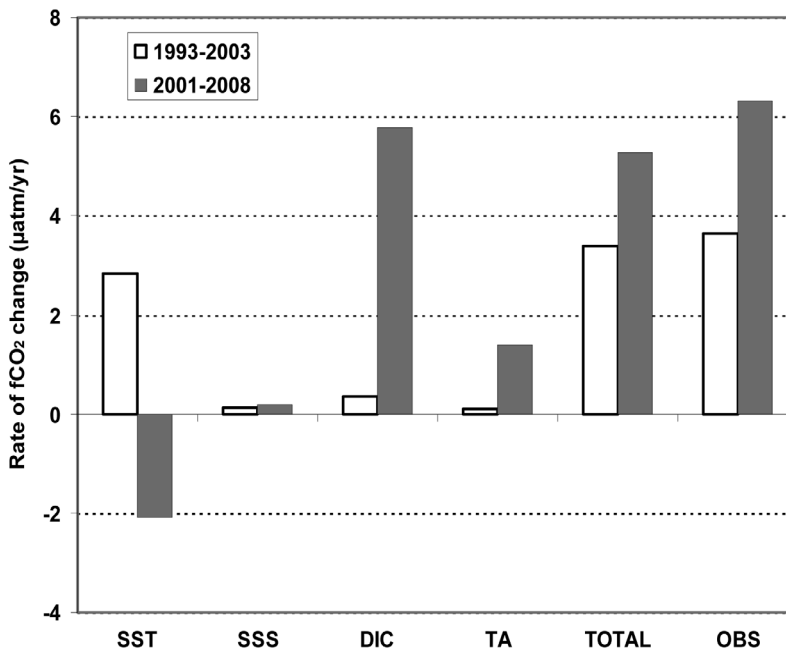
$\gamma =$  Revelle factor for DIC  $= (\partial \ln f\text{CO}_2/\partial \ln \text{DIC}) = (\partial f\text{CO}_2/\partial \text{DIC}) (\langle \text{DIC} \rangle/\langle f\text{CO}_2 \rangle)$ ;  $\gamma = 12$  for the nNASPG, and 11 for the sNASPG [*Chipman et al.*, 1993].

$\Gamma =$  Revelle factor of TA  $= (\partial \ln f\text{CO}_2/\partial \ln \text{TA}) = (\partial f\text{CO}_2/\partial \text{TA}) (\langle \text{TA} \rangle/\langle f\text{CO}_2 \rangle)$ ;  $\Gamma = -10$  for the nNASPG, and  $-9$  for the sNASPG.

$\eta$  and  $\Gamma$  were evaluated from “CO2SYS” [*Pierrot et al.*, 2006].

[22] The results of this partitioning process analysis were very similar in the nNASPG and sNASPG regions, therefore we calculated the mean of each term in equation (2) in the two regions and summarized the results in Figure 4. This





**Figure 4.** Decomposition of the changes in Sea Surface Temperature (SST), Sea Surface Salinity (SSS), Dissolved Inorganic Carbon (DIC) and Total Alkalinity (TA), on the rate of change of oceanic  $f\text{CO}_2^{\text{oc}}$  ( $\mu\text{atm yr}^{-1}$ ) in the North Atlantic Subpolar Gyre for two periods: winters 1993–2003 (white bars) and winters 2001–2008 (gray bars). The sum of all processes is also indicated (TOTAL) and compared to observed trends of  $f\text{CO}_2^{\text{oc}}$  (OBS) in winter for each period.

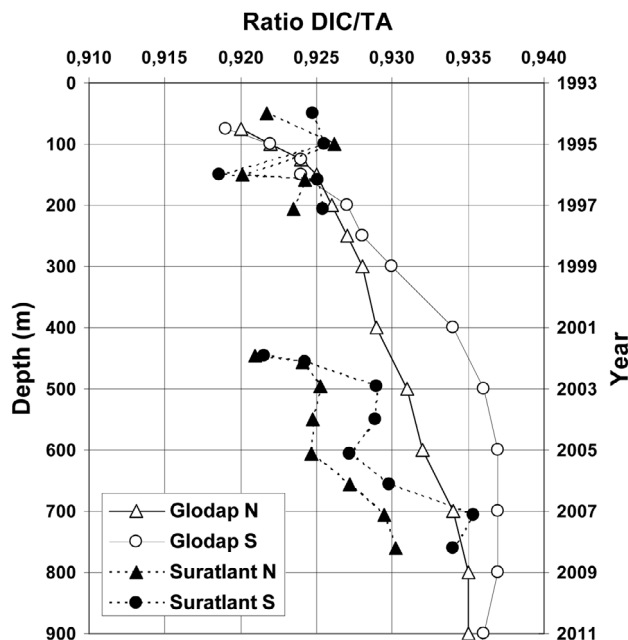
demonstrates how each of the four parameters affects  $f\text{CO}_2^{\text{oc}}$  either positively or negatively depending the period. The sum of all terms in equation (2) (TOTAL bars in Figure 4) agrees well with the observed  $f\text{CO}_2^{\text{oc}}$  trends also plotted (OBS bars in Figure 4). During the 2001–2008 period, the increase in DIC is identified as the major driver of the  $f\text{CO}_2^{\text{oc}}$  increase, while cooling and TA decrease represent small but together significant contribution. In contrast, during the 1993–2003 period, warming is the major cause for the  $f\text{CO}_2^{\text{oc}}$  increase.

[23] During 1993–2003 the  $f\text{CO}_2^{\text{oc}}$  trend was mainly controlled by warming associated with local heat flux anomalies [Häkkinen and Rhines, 2004] or the reduction of the subpolar gyre circulation and the advection of warm and saline southern waters into the Irminger Sea [Hátún et al., 2005]. The decrease in DIC expected from (i) reduced local mixing [Ullman et al., 2009] or (ii) advection of southern waters, was canceled by the anthropogenic  $\text{CO}_2$  increase [Corbière et al., 2007]. For the period 2001–2008, the changes in the chemical properties of the upper water of the NASPG and their impact on  $f\text{CO}_2^{\text{oc}}$  are clear, but the processes causing these changes have yet to be identified. All properties analyzed in this study (SST, SSS, nutrients, DIC and TA) have similar trends in the nNASPG and sNASPG, suggesting that they are related to large-scale processes and not to meso-scale structures such as eddies and frontal signatures occurring along or across the track.

[24] For the period 2001–2008, the salinity increase may be related to advection of water from temperate latitudes

(NAC) into the Irminger Sea as during the nineties, but this would not have induced the cooling of NASPG surface waters observed. Local decrease of freshwater flux would also affect SST (negative anomalies) and SSS (positive anomalies), but DIC and TA would vary in the same direction [e.g., Dore et al., 2003]. A possible explanation for both sea surface cooling and salinity increase could be the deeper mixing/convection observed in the Irminger Sea in 2007–2008 [Våge et al., 2008; Yashayaev and Loder, 2009]. The increase in nutrients, especially for nitrate, is also not consistent with horizontal advection as both southern or northern waters have lower concentrations than this part of the subpolar gyre. This is true also for DIC that demonstrates a well-defined maximum in the NASPG [Key et al., 2004] and therefore (as opposed to the period 1993–2003) advection of waters from the south or from the north cannot explain the rapid increase of DIC in recent years.

[25] As mentioned above, the observed nDIC trends in 2001–2008 in the NASPG (about  $1.6 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ , Table 1) are comparable to those observed in the surface waters of the Norwegian Sea (Ocean Weather Station M,  $66^\circ\text{N}/2^\circ\text{E}$ ) [Skjelvan et al., 2008], but higher than expected from anthropogenic  $\text{CO}_2$  uptake ( $<1 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ) [Olsen et al., 2006; Pérez et al., 2008]. When converting the increasing nutrient concentration to a change in DIC using the observed stoichiometric “Redfield” ratio of C:N (between 6.6 to 7.2 [Körtzinger et al., 2001]), we estimate DIC changes of between  $+2.3/+2.5 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  and  $+3.8/4.2 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  in the nNASPG and sNASPG respectively. Although



**Figure 5.** DIC/TA ratio as a function of depth for the northern and southern region in the NASG (mean of GLODAP data along Suratlant line, open symbols, left axis) and DIC/TA ratio as a function of time for the same regions (mean of Suratlant data, filled symbols, right axis). The DIC/TA ratio are higher in the south (in both data sets), and in recent years this ratio has increased in surface waters.

the derived DIC trends from nutrients are somewhat larger than the observed DIC trends (Table 1) this calculation is consistent with a more rapid increase of DIC in the southern region as observed. Note that adding the effect of TA variations (about  $-1 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ) would reduce DIC by about  $-0.3 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ . Thus the cumulated effect of both nutrients and TA would lead to a change for DIC of about  $+2.0/+2.2 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  and  $+3.5/3.9 \mu\text{mol kg}^{-1} \text{yr}^{-1}$  in the nNASPG and sNASPG respectively, in the range of the observed variations (Table 1). This indicates that the DIC and  $f\text{CO}_2^{\text{oc}}$  winter trends are not primarily driven by anthropogenic  $\text{CO}_2$  uptake, but more likely through vertical dynamical and/or biological processes, e.g., variations of remineralization in the upper layer.

[26] Given this close coupling between DIC and nutrient/TA trends, it is likely that processes such as vertical mixing and/or variations of remineralization in the upper layer have induced the DIC and nutrients (positive trend), and alkalinity changes (negative trend) and thus  $f\text{CO}_2^{\text{oc}}$  rapid increase. For the recent period (2007–2008) analysis of ARGO data suggests that convection/mixing increased dramatically during winter in the NASPG (up to 1000 m in the Irminger Sea during winter 2008) [Våge *et al.*, 2008]. This was also observed near Reykjanes Ridge in the nNASPG (V. Thierry, personal communication, 2009). This process would import more DIC and nutrients and decrease alkalinity (relative to DIC) in upper layers (in this region TA

is minimum at depth [Key *et al.*, 2004]). However, the change in convection is, at present, only well identified for 2007–2008, from hydrographic data, but not over the full period of this study (2001–2008). As a consequence, we cannot be certain that changes in mixing and convection explain the observed DIC, TA and  $f\text{CO}_2^{\text{oc}}$  changes over the whole period between 2001 and 2008. We note however that as both DIC and nutrients profiles have strong vertical gradients in the upper layers (0–1000 m), it is also possible that biogeochemical properties better reveal changes in vertical mixing than deduced from temperature and salinity alone.

[27] To investigate the mixing process hypothesis we have evaluated the temporal variations of the DIC/TA ratio in the nNASPG and sNASPG and compared these ratios with those deduced from water column observations (Figure 5). We first note that the DIC/TA ratios are generally higher in the sNASPG than in the nNASPG; this is true for both SURATLANT surface observations in 2003–2008 and for the mean DIC/TA ratio at depth between 200 and 900 m. Second, and in comparison with previous years, the DIC/TA ratios reveal clearly higher values in 2006–2008 in both regions. In 2007–2008, the DIC/TA was 0.93 in the nNASPG and 0.935 in the sNASPG; these values correspond to the DIC/TA ratio at depth around 400–500 m. If the vertical mixing is the main process explaining the temporal changes of DIC/TA (and nutrients), then surface waters with low DIC/TA ratio (about 0.92 for both regions on average) would have to be mixed with water much deeper than 500m. This is coherent with mixed-layers variations deduced from ARGO floats (up to 800–1000 m in winter 2007–2008) [Våge *et al.*, 2008]. The temporal variations of the DIC/TA ratios also suggest significant increase in winter 2003 and 2006 for both regions (Figure 5), that could also be interpreted as periods of deep winter mixing, although not clearly seen in hydrographic data. We note however, that in winter 2003 the location of  $4^\circ\text{C}$  isotherm recorded by ARGO floats in the Irminger Sea was relatively shallow, around 300 m compared to 500 m observed in 2002 and 2004 [Yashayaev and Loder, 2009].

[28] In the winter season, the primary production (PP) is low resulting from deep mixing and light limitation at northern high latitudes; consequently, a decrease of PP cannot explain the decadal increases of DIC and nutrients. This is consistent with satellite derived Chlorophyll-a that shows very constant concentrations in the NASPG during winter ( $0.2 \text{ mg m}^{-3}$ , in 1997–2008, from monthly SeaWiFS data at <http://oceancolor.gsfc.nasa.gov>). However, it is possible that changes in species composition in the North Atlantic (for both phytoplanktonic and zooplanktonic species, e.g., *Beaugrand* [2009]) could result in an increase of remineralization in upper layers (that would increase both nutrients and DIC, and decrease TA). Although changes of marine ecosystems (i.e., “systems more based on recycling and less on exportation,” *Beaugrand* [2009]) have been identified in the North Atlantic (and North Sea) in response to the NAO and ocean warming, their impact on the oceanic  $\text{CO}_2$  system has not yet been quantified. Also, successive blooms of coccolithophorids that were observed in the NA in 2003, 2004 and 2006 (*Brown and Yoder* [1994] and [http://cics.umd.edu/~chrisb/ehux\\_www.html](http://cics.umd.edu/~chrisb/ehux_www.html)) may also explain

large interannual variability of alkalinity; however, coccolithophorids blooms anomalies generally occur in the nNASPG, when we observed coincidental decrease in TA in both southern and northern regions. The coupling between slow variations or abrupt changes of marine ecosystems and oceanic carbon uptake merits further investigation. Regarding the ecosystems responses it is also important to note that the rapid  $f\text{CO}_2^{\text{oc}}$  increase observed in 2001–2008 leads to significant drop in pH, of  $-0.069 (\pm 0.007) \text{ decade}^{-1}$  (based on DIC/TA winter data and calculated following *Pierrot et al.* [2006]). This is four to six time faster than the change in pH (between  $-0.012$  and  $-0.02 \text{ decade}^{-1}$ ) observed in North Atlantic or North Pacific subtropical waters [*Santana-Casiano et al.*, 2007; *Dore et al.*, 2009].

## 5. Implication for the Ocean Carbon Uptake

[29] Previous data-based analysis concluded that it is not possible to clearly relate the variability of  $f\text{CO}_2^{\text{oc}}$  with NAO index [*Takahashi et al.*, 2009], mainly because of limited sampling frequencies in a region that experiences large  $f\text{CO}_2^{\text{oc}}$  variability (as also seen in Figures 3b and 3d). However, recent modeling studies do suggest that the surface ocean carbon cycle and hence the ocean carbon sink are influenced by the phase of the NAO [*Raynaud et al.*, 2006; *Thomas et al.*, 2008]. The positive NAO index in 2007 and 2008 would suggest that the gyre could return to 1990s conditions with a lag of  $\sim 2$  years [*Thomas et al.*, 2008].

[30] In our study the regional-scale consistency of the trends are observed for all properties in 1993–2003 and 2001–2008, in both the sNASPG and nNASPG regions (Table 1). These results support the conclusion that the sea surface carbon system variability is governed by basin scale variations. These may be potentially linked to the NAO, particularly the positive-to-negative shifts that occurred in 1995–1996 and 2000–2001 (Figure 3). It is also possible that over the investigated period, 1993–2008, the acceleration of  $f\text{CO}_2^{\text{oc}}$  growth rate may be related to the AMV index that has been in a positive phase since 1995 [*Ting et al.*, 2009; *Reverdin*, 2010]. The rapid  $f\text{CO}_2^{\text{oc}}$  trends observed since 2001 might be a manifestation of a regime shift rather than a long-term trend, although it is not possible from our data to predict the future response based solely on NAO phase as has been suggested by *Thomas et al.* [2008].

[31] In comparison with other studies, the  $f\text{CO}_2^{\text{oc}}$  trend of  $3.7 (\pm 0.6) \mu\text{atm yr}^{-1}$  estimated between 1993 and 2003, is at the upper bound of those prior to 2006, spanning a range of  $1.6$ – $3.8 \mu\text{atm yr}^{-1}$  [*Lefèvre et al.*, 2004; *Omar and Olsen*, 2006; *Corbière et al.*, 2007; *Schuster et al.*, 2009; *Takahashi et al.*, 2009]. The lower value,  $+1.6 (\pm 0.3) \mu\text{atm yr}^{-1}$ , corresponds to the mean of the  $f\text{CO}_2^{\text{oc}}$  trends estimated north  $45^\circ\text{N}$  for a longer period, covering 1972–2006 [*Takahashi et al.*, 2009], as well as the lower bound of the estimates of *Omar and Olsen* [2006] for the subpolar gyre between 1972 and 1990. The maximum value of  $3.8 (\pm 0.8) \mu\text{atm yr}^{-1}$  corresponds to the mean of the  $f\text{CO}_2^{\text{oc}}$  trend north of  $50^\circ\text{N}$  based on data for the recent period, 1990–2005 [*Schuster et al.*, 2009] as well as the upper bound presented by *Omar and Olsen* [2006]. The difference could be explained by the period length and/or by an acceleration of the

$f\text{CO}_2^{\text{oc}}$  trend in recent years as suggested by this study when analyzing the later period 2001–2008, which displays  $f\text{CO}_2^{\text{oc}}$  trends  $> 5 \mu\text{atm yr}^{-1}$  (up to  $7 \mu\text{atm yr}^{-1}$ ). This is an unprecedented  $f\text{CO}_2^{\text{oc}}$  growth rate in the North Atlantic or any other oceanic region and results in the NASPG acting as a source of  $\text{CO}_2$  for the atmosphere in the wintertime during this period.

[32] In this study we focused on winter trends as summer  $f\text{CO}_2^{\text{oc}}$  exhibits large small-scale space-time heterogeneity that may bias results when discussing the decadal trends. Summer observations however also indicate (both calculated and measured  $f\text{CO}_2^{\text{oc}}$ ) that  $f\text{CO}_2^{\text{oc}}$  was higher in recent years (Figures 3b and 3d). This is consistent with a previous analysis that took into account summer observations (but over a longer period), and concluded that summertime  $f\text{CO}_2^{\text{oc}}$  increased at a rate close to, or higher than in the atmosphere in the NASPG [e.g., *Corbière et al.*, 2007] ( $+1.8 \mu\text{atm yr}^{-1}$  in 1981–2001). Therefore, with the rapid growth rate estimated in winter 2001–2008 and without an apparent compensating negative trend in summer, we conclude that on an annual basis, the ocean carbon uptake in the NASPG has likely been reduced more rapidly in 2001–2008 compared to previous decades. This scenario is important, but cannot be applied a priori, to the whole North Atlantic basin without further investigation and confirmation, e.g., using comprehensive  $f\text{CO}_2^{\text{oc}}$  data synthesis such as the SOCAT international project [*IOCCP*, 2008].

## 6. Summary

[33] The measurements obtained since 1993 from the SURATLANT program provide a comprehensive view of the decadal evolution of the NASPG carbon sink. The winter observations show that since 1993  $f\text{CO}_2^{\text{oc}}$  has increased at a rate faster than the atmospheric  $\text{CO}_2$ , leading to a reduction of the ocean carbon uptake. Over the most recent period (2001–2008), the observations indicate an  $f\text{CO}_2^{\text{oc}}$  trend of between  $+5.5$  and  $+7.2 \mu\text{atm yr}^{-1}$  almost twice that observed between 1993 and 2003 ( $+3.6 \mu\text{atm yr}^{-1}$ ). The increase of  $f\text{CO}_2^{\text{oc}}$  since 2001, in contrast to the earlier period 1993–2003 which is driven by changes in SST [*Corbière et al.*, 2007; *Schuster et al.*, 2009], is primarily ( $>95\%$ ) explained by changes in seawater chemistry. The recent increase of convective processes/vertical mixing in the NASPG may explain these changes and is broadly consistent with observed changes of the DIC/TA ratio. However, we cannot exclude biological processes, e.g., changes in remineralization that could occur in relation to variations of marine species composition. A conclusion on dynamical and/or biological processes that impact on the carbon cycle in surface waters is not reached or quantified in our analysis, but needs to be investigated in future studies. Another important link between ocean  $\text{CO}_2$  variability and ecosystems response concerns the diminution of pH (so-called ocean acidification) associated to the fast increase of surface  $f\text{CO}_2^{\text{oc}}$ , here deduced from DIC and TA data. In 2001–2008 we estimate a pH decrease of  $-0.069 (\pm 0.007) \text{ decade}^{-1}$ , much faster than what was observed in oligotrophic subtropical waters in the North Atlantic and Pacific oceans [*Santana-Casiano et al.*, 2007; *Dore et al.*, 2009].

[34] The diminution of the ocean carbon sink (or its stability) has been also observed in several regions, especially at high latitudes, in the Southern Ocean [Le Quéré et al., 2007; Metzl, 2009]. Other investigations of the  $f\text{CO}_2^{\text{oc}}$  trends in the ocean do not show an increase of the carbon sink (except the Bering and Okhotsk Seas [Takahashi et al., 2006]). Our results, in conjunction with other data-based analyses, could explain the reduction of the ocean carbon uptake since about 15–20 years and its impact on the air-borne fraction [Canadell et al., 2007]. If the decrease of the oceanic uptake is confirmed in other regions then, in the near future, atmospheric  $\text{CO}_2$  levels would rise faster with potential positive feedback on climate change.

[35] The results presented and discussed in this work highlight the need for continued long-term sea surface ocean observations of carbon properties (DIC, TA and  $f\text{CO}_2$ ) in the framework of considering sampling strategies [e.g., Lenton et al., 2009; Monteiro et al., 2009] and investigation into the decadal variability at the scale of the North Atlantic [Gruber, 2009; Watson et al., 2009]. It is important to continue ocean observations, which document the relative importance of the decadal NAO/AMV effects and superimposed chemical changes that intensified the rapid reduction in the North Atlantic  $\text{CO}_2$  sink flux and possibly accelerate the acidification of surface waters. Estimating and understanding better the rapid change observed in ocean  $\text{CO}_2$  system and its potential impact and feedback on the ocean carbon sink and marine ecosystems represent also an important and significant challenge for both atmospheric and oceanic models.

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