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Abstract

A biogeochemical general circulation model is used to assess the impact of climate variability from 1992-2006 on air-sea CO₂ fluxes and ocean surface pCO₂ in the North Atlantic and to understand trends in the North Atlantic carbon sink over this time period. Results show a strong correlation between flux and pCO₂ variability. Consistent with observations, the model output indicates large changes in the physical and chemical systems of the basin. An analysis of the trends of dissolved inorganic carbon (DIC), alkalinity (ALK), and sea-surface temperature (SST), combined with model-derived DIC tendency terms allow for an investigation of the mechanisms that dominate the spatial variability and magnitude of the trends in the air-sea fluxes and pCO₂. Subpolar trends in pCO₂ are driven dynamically, primarily through changing vertical supply of DIC. Subtropical trends are instead controlled primarily by changes in sea surface temperature. The amplitude of the ocean pCO₂ and air-sea flux trends are largely related to the increase in atmospheric CO₂, but changes to the forcing and circulation of the North Atlantic during this period set the spatial patterns.
1. INTRODUCTION

Since the Industrial Revolution, there has been a marked increase in the concentration of CO₂ in the atmosphere. Human activities, including the burning of fossil fuels, the production of cement, and changes in land use have led to a change in atmospheric CO₂ concentrations from about 280 ppm in 1850 to values of about 380 ppm in 2007 (Keeling & Whorf, 2005). This increase, however, has been moderated through terrestrial and oceanic sinks that have assimilated roughly half of the total CO₂ emitted from anthropogenic sources (IPCC, 2007).

Changes in land use and biomass burning have led to a terrestrial flux of CO₂ from the land to the atmosphere, such that the oceans have served as the only “true” sink for anthropogenic CO₂ over the past 200 years, and were it not for oceanic uptake, atmospheric CO₂ would be 55 ppm higher than present levels (Sabine et al., 2004).

The growth rate of atmospheric CO₂ varies considerably more than estimated anthropogenic CO₂ emissions (Conway et al., 1994; Peylin et al., 2005), indicating that interannual variability of terrestrial and ocean CO₂ sinks are important. Year-to-year changes in climate forcing can have profound impacts on ocean temperature, chemistry, and circulation, all of which impact the ocean carbon cycle (McKinley et al., 2004a). Aside from the prominent role of ENSO variability, the causes of interannual variability in the mid to high latitude ocean sink is poorly understood, with estimates of sink magnitudes and variability largely inconsistent (McKinley et al., 2004a; Gurney et al., 2002; Keeling et al., 1996; Peylin et al., 2005; Baker et al., 2006), though increased agreement between the methodologies is found when a smaller grid is used for the inversion (Rödenbeck et al, 2003; McKinley et al., 2004b).

There is also significant spatial variability in the ocean sink. Due to differences in ocean circulation and chemistry, each basin has its own inherent ability to absorb atmospheric CO₂.
The Atlantic Ocean, in particular the North Atlantic, plays a significant role in the uptake of anthropogenic CO$_2$. Takahashi et al. (2002) estimated that the Atlantic accounts for 41% of the global flux of CO$_2$ into the ocean. The northern basin comprises only 15% of the global ocean surface, but has absorbed 23% of the anthropogenic carbon stored in the oceans (Sabine et al. 2004).

However, there have been significant changes in the North Atlantic over the last few decades. Schuster and Watson (2007) report on volunteer observation ship (VOS) track data between the Caribbean and the U.K. Along this track, they report a 50% decrease in the sink of atmospheric CO$_2$ from 1994/95 to 2002/2005 in the northeast area of the section. This may be related to decreasing geostrophic velocity in the subpolar gyre since 1994 (Häkkinen & Rhines, 2004). In this paper, we use an ocean biogeochemical model to study the spatial nature of the changes to the chemical and physical properties to better understand the North Atlantic’s shifting ability to take up CO$_2$.

2. METHODS

2.1 The Physical-Biogeochemical-Ecosystem Model

The MIT Ocean General Circulation Model (Marshall et al., 1997a,b) was regionally configured for the North Atlantic between 20°S and 81.5°N, with a horizontal resolution of 0.5° latitude and 0.5° longitude. The model was set up to have 23 vertical levels with a resolution of 10 m thickness at the surface increasing to 500 m thickness for depths greater than 2200 m. The Gent-McWilliams (Gent and McWilliams, 1990) eddy parameterization and the KPP boundary layer mixing scheme (Large et al., 1994) were employed to represent sub-gridscale processes.
general circulation model was forced with daily fields from the NCEP/NCAR Reanalysis I
(Kalnay et al., 1996) for 1992-2006.

A sponge layer, similar to Williams et al. (2006), was included along regional boundaries
to dampen the accumulation of tracers in regions that would in reality allow for the flow of
tracers to and from other ocean basins. Along the southern boundary, temperature, salinity,
dissolved inorganic carbon (DIC), and nutrients were relaxed to climatology. At the Strait of
Gibraltar, temperature and salinity alone were relaxed to climatology, as Béthoux et al. (1998)
suggest that nutrient inflow from the Mediterranean is insignificant. A sponge layer at the
northern boundary (north of 81.5°N) was discarded after tests showed that such a sponge layer
was inconsequential, in this configuration, to the regional circulation, temperature, and salinity.

The pelagic ecosystem is parameterized with one zooplankton class and two phytoplankton
classes: diatoms and “small” phytoplankton (Dutkiewicz et al., 2005). Carbonate chemistry is
modeled following Follows et al. (2006). The cycling of carbon, alkalinity, phosphorus, silica
and iron are explicitly included in the biogeochemical model (see Dutkiewicz et al., 2005 and
Bennington et al., 2008 for a more detailed description). Atmospheric pCO₂, including a seasonal
cycle, was taken from Mauna Loa observations (Keeling et al., 2005). The flux of carbon
dioxide between the ocean and atmosphere was parameterized following Wanninkhof (1992) and
with updated coefficients based on the ¹⁴C inversion of Sweeney et al. (2007). We define a
positive flux as one directed into the ocean.

The physical model was spun up for 81 years while relaxing sponge layer temperature
and salinity to monthly climatology (World Ocean Atlas [WOA], Locarnini et al., 2006; Antonov
et al., 2006) with timescale of 2 and 4 weeks, respectively. SST was relaxed to 1992-2006
climatological satellite-based estimates (Reynolds et al., 2007). After the physical spin up, the
biogeochemical model was initialized with GLODAP (Key et al., 2004) DIC and ALK climatology, WOA nutrients and oxygen (Garcia et al., 2006a, 2006b), low values of phytoplankton and zooplankton, and atmospheric pCO$_2$ fixed at 356 ppm (roughly the 1992 level). The biogeochemical model was spun up for 70 additional years until all major drift in biogeochemical parameters was eliminated. The results discussed here come from an additional 15 years using the full biogeochemical model with 1992-2006 NCEP daily forcings, a time-varying atmospheric pCO$_2$ field, and relaxation to monthly-varying SST (Reynolds et al. 2007).

To separate the effects on the trends due to changes in climate forcing from the effects on the trends due to anthropogenic increase in atmospheric CO$_2$, a second run of the model was conducted to assess the effects of the same climate forcings on a carbon system with a constant preindustrial atmospheric CO$_2$ concentration. Starting from the same physical spin up from above, the biogeochemical model was initialized with GLODAP estimates of preindustrial DIC and ALK climatology (Key et al., 2004) and a constant atmospheric CO$_2$ of 280 ppm. This was run until levels of DIC and ALK stabilized within the model. Once the carbon and other biogeochemical parameters stabilized, the model was run with the same interannual varying daily NCEP forcings for 1992-2006 with the atmospheric CO$_2$ remaining fixed at 280 ppm.

### 2.2 Post-Simulation Methods

As described by Takahashi et al. (1993), pCO$_2$ of the surface ocean can be separated into influences from DIC, Temperature (T), ALK, Salinity (S), Phosphate (PO4), and Silicate (SIL) according to the following equation:

$$ \frac{dpCO_2}{dt} = \frac{\partial pCO_2}{\partial DIC} \frac{dDIC}{dt} + \frac{\partial pCO_2}{\partial T} \frac{dT}{dt} + \frac{\partial pCO_2}{\partial T} \frac{dALK}{dt} + \frac{\partial pCO_2}{\partial S} \frac{dS}{dt} + \frac{\partial pCO_2}{\partial PO4} \frac{dPO4}{dt} + \frac{\partial pCO_2}{\partial SIL} \frac{dSIL}{dt}. $$
Each component was estimated by calculating the pCO$_2$ with the carbonate chemistry equilibrium constants of the model (Follows et al., 2006; Merbach et al., 1973; Dickson, 1990 in Dickson and Goyet, 1994), using the deseasonalized variability of the component of interest and setting the values of the remaining parameters to their long-term means (LeQuéré et al., 2003; McKinley et al., 2004a, 2006). The sum of each of the components matches reasonably well with the total pCO$_2$ provided daily model output is used. Salinity, Phosphate, and Silicate represented a small proportion of the overall variability of the total pCO$_2$ and are therefore excluded from subsequent analysis.

As will be discussed later, the model output includes the change in DIC over time due to a number of individual mechanisms driving the DIC concentration. These include effects from vertical mixing, biological uptake, remineralization, freshwater influence (dilution/concentration from precipitation/evaporation), horizontal advection, air-sea flux, and storage (total change over time). We call these tendency terms “DIC diagnostics.” The averages of each of these terms was calculated over the top 100m of the model and are used to assess the changes in DIC within a surface layer indicative of a well-mixed surface layer where most of the biological productivity occurs.

3. RESULTS

3.1 Model/Data Comparison

For the most part the physical model compares favorably with what limited observations are available in the North Atlantic. Modeled Gulf Stream transports are within 70% of those observed at Cape Hatteras (Tomczak and Godfrey, 1994) and in the Florida Strait (Cunningham et al., 2007), as expected for an OGCM of this resolution. Model mixed layer depth (MLD)
compared reasonably well with that of WOA climatology, though the model sometimes
overestimating maximum winter MLDs at high latitudes. We define the model MLD as the
depth where $\sigma_T$ is different from that of the surface by 0.125 kg/m$^3$ or more (Levitus, 1982).

As shown by Takahashi et al. (1993), ocean surface pCO$_2$ values can be separated into
temperature-driven and non-temperature-driven components. Bennington et al. (2008) show that
modeled seasonal differences of pCO$_2$ and related temperature and non-temperature components
compare favorably with climatological observations (Takahashi et al., 2002). The model does
reasonably well in capturing the mean seasonal cycle of pCO$_2$ and its components (Bennington et
al., 2008), a challenging task as illustrated by a recent model/data intercomparison in the North
Pacific (McKinley et al., 2006).

A valuable dataset for comparing the interannual variability in the data with observations
is available from the Bermuda Atlantic Time series Station (BATS, 31°40’N, 64°10’W, [Bates,
2007]), providing monthly-resolved Conductivity-Temperature-Depth (CTD) instrument data
and carbon measurements since 1988. The modeled pCO$_2$ compares favorably with observations
in both the timing and magnitude of the pCO$_2$ cycle (figure 1a). Similarly, the model captures
the SST variability seen in the observations (figure 1b). Modeled DIC matches the timing of the
seasonal cycle (figure 1c) but underestimates summer DIC drawdown because modeled
summertime productivity is too low (Bennington et al., 2008). Consistent with observations
(Bates, 2007), modeled pCO$_2$ variability at BATS is mostly driven by variability of SST.

At BATS, Bates (2007) finds seasonal detrended surface DIC and ALK increases of 1.27
± 0.08 μmoles kg$^{-1}$ yr$^{-1}$ and 0.58 ± 0.09 μmoles kg$^{-1}$ yr$^{-1}$, respectively, during the 1988-2003
period, and an increase in oceanic pCO$_2$ of 1.80 ± 0.13 μatm yr$^{-1}$ during the 1984-2005 period.
For 1992-2006, this model puts such increases at 1.08 μmoles kg$^{-1}$ yr$^{-1}$, 0.33 μmoles kg$^{-1}$ yr$^{-1}$,
and 0.71 μatm yr⁻¹, respectively. While the trend dates for these analyses are not exactly the same, the model captures the appropriate trends in this region, though underestimates their absolute magnitude on a per year basis. Differences in trends may be due to the differences in the time periods of analysis.

3.2 Temperature versus Dynamical Controls on Surface pCO₂

Consistent with the findings of LeQuéré et al. (2003), the two major physical controls on surface ocean pCO₂ are temperature and dynamical mixing, and their effects are typically in opposition. For example, increased mixing brings older DIC-rich waters to the surface to drive an increase in pCO₂, but this increased mixing is also related to lower SST, driving a decrease in the pCO₂. Much of pCO₂ variability is dominated by one these controls (temperature vs. dynamics), as can be seen from the correlation of pCO₂ and SST (figure 2a). Regions exhibiting a negative correlation between pCO₂ and SST are considered “dynamics-driven,” such that a decrease in SST drives an increase in mixing, bringing more DIC to the surface, and thus increasing the pCO₂. Regions exhibiting a positive correlation are “temperature-driven,” such that an increase in SST directly forces an increase in pCO₂ through the thermodynamic controls on gases in seawater. As seen in figure 2a, the dynamics-driven controls on pCO₂ are confined to the subpolar gyre, and temperature-driven controls dominate the entire basin south of 45°N. Such a spatial distribution in the controls on pCO₂ is consistent with climatological observations (Takahashi et al., 2002).

That the carbon cycle is driven by dynamics in the subpolar gyre is consistent with the deep mixing in this region: the high concentration of DIC mixed to the surface more than offsets the solubility effects due to changes in temperature (Takahashi et al., 2002). Figure 2b shows that
the subpolar pCO\textsubscript{2} is indeed positively correlated with deeper mixing. Observations from Lueger et al. (2008) confirm the positive correlation between pCO\textsubscript{2} and MLD in the subpolar gyre and negative correlation in the subtropical gyre.

3.3 Spatial variability and assessment of trend

The main mode of observed physical variability in this region is the North Atlantic Oscillation (NAO; Hurrell et al., 2003). Thomas et al. (2008) have considered the influence of NAO changes on the North Atlantic pCO\textsubscript{2} and CO\textsubscript{2} flux for 1979-2004 and find some intriguing relationships. However the model results here, consistent with Thomas et al. (2008), show that the correlation between the timeseries of air-sea CO\textsubscript{2} fluxes and the NAO is weak or non-existent at the basin or gyre scale for 1992-2006, and thus it is not a focus for our analysis. Instead, we focus on the large-scale trends.

There is a strong trend in regional air-sea CO\textsubscript{2} fluxes (figure 3a). The magnitudes of the modeled regional fluxes are consistent with 1995 observations (Takahashi et al., 2002). There is interannual variability, but it is superimposed onto the increasing trend. The regional CO\textsubscript{2} fluxes from the preindustrial run (thin lines, figure 3a) shows similar interannual variability to that of the modern run, but the overall trend is missing. We conclude that the air-sea flux trend over this period is due to the increasing atmospheric CO\textsubscript{2} trend and not to long-term variability in the physical climate or biogeochemistry.

To examine the regional patterns of the modeled trend in the CO\textsubscript{2} flux, we consider the difference between 4-year means at the start and end of the run (2003-2006 mean minus 1992-1995 mean). (Different years/lengths for the means were considered and showed the same qualitative results). The trend in the CO\textsubscript{2} flux (figure 3b) shows a “quad-pole” of 4 localized
centers of action with an increase in the flux in the western North Atlantic and in the subpolar gyre south of Greenland. The air-sea flux shows decreases in the eastern side of the basin, off the coast of the Iberian Peninsula and further south in the subtropical gyre.

To assess the main modes of variability within the modeled carbon system, a principal component analysis (PCA) was conducted on the pCO$_2$ and CO$_2$ flux output over 1992-2006. The first empirical orthogonal functions (EOF) of pCO$_2$ and CO$_2$ flux explain 73% and 36% of the overall variance, respectively, and both show a strong trend as seen in the first principal component (PC1, figure 4). The first EOF of the pCO$_2$ (figure 4a) is positive over the entire basin, and the related increase in the first PC (figure 4c) indicates that the positive spatial pattern intensifies over time. The first EOF of the CO$_2$ flux (figure 4b) is positive over the majority of the basin with values up to 500 mmol/m$^2$/yr in the subpolar gyre, and some negative values as low as -50 mmol/m$^2$/yr (sea to air) in the subtropics extending up along the eastern boundary. These negative regions are collocated with areas of largest positive pCO$_2$ trend seen in the plot of the first EOF (figure 4a).

The trends in the PC1 of pCO$_2$ and the PC1 of the air-sea flux (figure 4c) are highly correlated ($r=0.87$), indicating a strong relationship between the variability of the pCO$_2$ and the flux. The spatial patterns are also similar (figure 4a, b). The high variance explained by each EOF1 and the similarity in patterns between the EOFs and the difference plots (Fig. 3b) suggest that a high proportion of the overall changes are driven by the trend. Additionally, in light of the similarity of such North Atlantic trends between these model results and in comparable observations (Schuster and Watson, 2007), this paper will focus on the trend itself to understand regional mechanisms. We elaborate further on the model/data comparison in the Discussion section.
Because \( p\text{CO}_2 \) variability is so strongly related to the \( \text{CO}_2 \) flux variability, we will use \( p\text{CO}_2 \) to analyze the carbon system over time. The trend in \( p\text{CO}_2 \) (figure 5a) shows an increase over the entire basin, but this increase is spatially variable, with a quad-pole pattern of lesser/greater increase similar to that described above. The overall positive trend in \( p\text{CO}_2 \) is driven mostly by the increase in atmospheric \( p\text{CO}_2 \) (figure 3a). The spatial differences in the magnitude of the ocean \( p\text{CO}_2 \) increase are related to a number of other factors that influence the chemical balance of carbon in the ocean.

The trends in the \( p\text{CO}_2\text{-ALK} \), \( p\text{CO}_2\text{-DIC} \), and \( p\text{CO}_2\text{-SST} \) components (Eq.1) are also shown in figure 5. The range of variability in the trends of the components is large (±200 \( \mu\text{atm} \)) relative to that of the total \( p\text{CO}_2 \) (±30 \( \mu\text{atm} \)), indicating that the components largely counteract each other. While the spatial variability in the total \( p\text{CO}_2 \) trend encompasses much of basin, high variability in the trend of the components is confined to the subpolar gyre. Large increases in \( p\text{CO}_2\text{-ALK} \) and \( p\text{CO}_2\text{-SST} \) are only in part balanced by large decreases in \( p\text{CO}_2\text{-DIC} \) in the subpolar gyre. Though analysis methodologies differ, these modeled trends in the \( p\text{CO}_2 \) components are broadly consistent with those found by Thomas et al. (2008) for the mid-1990’s to the mid-2000’s.

While the overall magnitude in the modeled western subtropical \( p\text{CO}_2 \) trend may be slightly low as compared to observations at BATS (see section 3.1), the spatial variability captures the tri-pole of subtropical observations in the trend (Schuster and Watson, 2007) quite well, with larger increases in \( p\text{CO}_2 \) in the eastern basin, both north and south, separated by smaller increases in the west that extend east over the central subtropics. This spatial pattern in the subtropics is driven mostly by changes in SST, as seen from the trend in the \( p\text{CO}_2\text{-SST} \) component (figure 5d), with decreases in the west and increases in the east. The \( p\text{CO}_2\text{-ALK} \) and
pCO\textsubscript{2}-DIC components (figures 5b and 5c) oppose each other throughout most of the region, though the DIC component is slightly larger and leads the net increase in pCO\textsubscript{2}.

The spatial change in pCO\textsubscript{2}-SST is highly consistent with observations, including the warming in the Northwest (ICES, 2006) from 1992-2006. Similarly, satellite data from Reynolds et al. (2007) show increased SST north of 50N over the same time period. These changes in SST have a direct impact on the surface pCO\textsubscript{2}, as evidenced from the trend in the pCO\textsubscript{2}-SST component (figure 5d).

### 3.4 Diagnostics of the DIC Tendency Terms

To further the understanding of DIC changes, the model “DIC Diagnostics” (see section 2.2) allow quantification of the mechanisms driving the trends in DIC concentrations: DIC-vertical, DIC-biology, DIC-fresh, DIC-horizontal, and DIC-flux. Each of these terms quantifies the rate of change in DIC due to each mechanism, described below (units of mmol/m\textsuperscript{3}/yr). The mean and difference plots from the two time periods for each of these DIC diagnostics is shown in figure 6.

DIC-vertical (figures 6a) quantifies the change in DIC due to vertical advection, diffusion, and all mixing processes. Regions with strong vertical supply of DIC occur along the Gulf Stream and into the subpolar gyre where strong currents and deep mixed layers entrain DIC to the surface. These same regions also show the greatest change from 1992/95 to 2003/06 with a strong decrease in the vertical supply of DIC to the surface. These results are consistent with the decreases in MLD seen in this region (figure 7a). The largest decrease in MLD occurs in the Labrador Sea and off the southern edge of Greenland and is consistent with observations (Lazier et al., 2002; Yashayaev, 2007). Not surprisingly, shallower MLDs appear to drive a decrease in
vertical supply, thus mixing fewer nutrients to the surface (figure 7b). In the subtropics, DIC vertical supply is rather low, with small or non-existent trends.

DIC-biology (figures 6b) quantifies the loss of DIC due to uptake of DIC by photosynthesis: negative DIC-biology is indicative of enhanced productivity. Highest biological uptake occurs in subpolar regions. Differences in DIC-biology from 1992/95 to 2003/06 are rather small, with some strong decrease in biological uptake along 45N. The model also shows some increase in the uptake in the western subtropics along 30N, which indicates of a southward shift in the interface between high and low productivity.

DIC-fresh (figures 6c) quantifies the change in the concentration of DIC due to changes in the input of freshwater: negative DIC-fresh indicates a freshening of surface waters and dilution of DIC. In the mean there is dilution of DIC (more precipitation than evaporation) in the high north and along the equator, and a concentration (more evaporation than precipitation) elsewhere in the basin. The modeled trend of concentrating DIC in the subpolar gyre and Labrador Sea is consistent with recent observations of increased salinity in this region (ICES, 2006; Yashayaev, 2007).

DIC-horizontal (figure 6d) quantifies the change in DIC due to horizontal advection and diffusion. Not surprisingly, the regions of strongest mean DIC-horizontal occur along the Gulf Stream. The DIC-vertical diagnostic suggests that large amounts of DIC are brought to the surface along the Gulf Stream, but this DIC is rapidly advected out of the region (the strong negative values in DIC-horizontal). Trends of this term in the northeast may be related to changes in the path of the Gulf Stream, consistent with the reduction in subpolar gyre strength over the time period (Häkkinen & Rhines, 2004).
DIC-flux (figure 6e) quantifies the change in DIC due to the air to sea flux. Much of the mean addition of DIC from the air-sea flux is driven by the withdrawal of DIC from the water by biological productivity: the regions of strong DIC-biology and DIC-flux are highly collocated. Trends in the DIC-flux, however, appear to be less related to biological changes but more to changes in the vertical supply. Large subpolar increases in the air-sea flux appear to be driven by the strong decrease of supply of DIC due to vertical processes. The slow rise in atmospheric pCO$_2$, coupled with the spatial variability of the surface ocean pCO$_2$ due to combined effects of SST, DIC, and ALK (figure 5), allows for the air-sea ΔpCO$_2$ to grow with time (figure 5a) and thus a significant flux trend to occur.

3.5 Preindustrial Trend

The preindustrial run uses the same internannual forcing as the “anthropogenic” run, but without the increasing atmospheric pCO$_2$. As such, this run helps us identify the changes to pCO$_2$ and air-sea CO$_2$ flux that come from circulation changes only. The pCO$_2$ trend for the preindustrial run (figure 8a) shows similar spatial patterns, but considerably lower values than those found in the anthropogenic pCO$_2$ trend (figure 5a). The regions in the anthropogenic run that demonstrate less increase in pCO$_2$ (subpolar gyre, western basin along 30N) actually show a decrease in pCO$_2$ for the preindustrial run. This comparison illustrates that natural variability in the climate forcing over the region is driving the spatial variability of the surface pCO$_2$, consistent with the results of Thomas et al. (2008).

While the preindustrial trends in the ALK (figure 8b) and SST (figure 8d) are nearly identical to the trends in the anthropogenic run, the preindustrial pCO$_2$-DIC trend, however, shows a much stronger and broader (spatially) decline in the subpolar gyre. This component is
directly connected to the different atmospheric pCO$_2$ forcing in this run. The preindustrial pCO$_2$-DIC trend matches the shoaling trend in the MLDs (figure 7a), confirming that reduced mixing due to changes in climate forcing is driving the decrease in DIC in this region. In the Eastern Subpolar region of the anthropogenic run, the increased air-sea flux of CO$_2$, due to the atmospheric pCO$_2$, compensates for the reduced vertical supply, but this flux is not strong enough to fully compensate in the west.

4. Discussion

In our model study, the dynamical control of pCO$_2$ in the subpolar regions is largely driven by changes to the DIC concentration. While changes in biological productivity, dilution/concentration by freshwater, and horizontal advection alter DIC on a local level, changes in mixing and vertical supply have the largest impact on the concentration of DIC in the top 100m across the subpolar gyre. From 1992-2006, the model indicates that subpolar DIC decreases are due to a decrease in vertical supply. The air-sea CO$_2$ flux responds to the strong decreases in ocean surface pCO$_2$. Declining subpolar mixing from 1992-2006 also drives changes in alkalinity. With less nutrients coming to the surface, there is a reduction in surface alkalinity, which drives the strong increase in pCO$_2$-ALK (figure 5b) in this subpolar gyre. This effect, however, is weaker than pCO$_2$-DIC changes. Based on a qualitative analysis of temporal trends in observed pCO$_2$ and its components, Thomas et al. (2008) suggest NAO-forced changes in horizontal processes dominate over the North Atlantic pCO$_2$ variability. However, Thomas et al. (2008) do not analyze changes in vertical mixing. Our analysis suggests that though horizontal processes do redistribute DIC, these effects are relatively localized, and changes in vertical processes are more important to the large-scale trends in the subpolar gyre.
In the subtropics, the spatial variability of pCO$_2$ is mostly controlled by variability of SST. The small increases in overall pCO$_2$ due to pCO$_2$-ALK and pCO$_2$-DIC (figure 5a) in the western subtropical gyre are limited by the decreases in pCO$_2$-SST (figure 5d). In the eastern subtropical gyre, where the pCO$_2$-SST is positive, pCO$_2$ shows a larger increase from 1992-2006.

Schuster and Watson (2007), using in situ measurements of pCO$_2$, suggest a spatially varying change in the air-sea flux of CO$_2$ from the mid-1990s to early-2000’s: a decreasing flux in the central subtropical gyre, the eastern subtropics near the Iberian Peninsula, and the western subpolar gyre, and an increasing flux in the western subtropical gyre (Schuster and Watson, 2007, figure 6). While the magnitude of the changes to the air-sea CO$_2$ flux in the model is smaller than that of the observations, the spatial distribution of the increase/decrease is similar to the observations, except in the western subpolar gyre (Figure 4b). Corbière et al. (2007, summarized in Watson and Schuster, 2007) observe a reduction of the sink in this subpolar gyre region. The limited observations they consider suggest that DIC was relatively constant over the time period, and Corbière et al. (2007) argue that the increase in the subpolar pCO$_2$ is driven by the increase in temperature since the mid-1990s. Our model suggests that while SSTs have indeed warmed, the pCO$_2$ change has been dominated by a marked decrease in DIC in this region, driven by decreases in mixing and related vertical supply of DIC.

Decreased mixing in the western subpolar region has also been observed by other studies. Hydrographic observations in the Labrador Sea suggest reductions in MLDs due to a reduction in wintertime convection (Lazier et al., 2002; Yashayaev, 2007). Similarly, Häkkinen and Rhines (2004) have reported on a weakening of the subpolar gyre through the late 1990s related to reduced heat fluxes. Such a spin down of the gyre should lead to a declining slope of the
isopycnals and subsequent stratification. An analysis of the changes in the barotropic stream function and tracer release simulations with this model have shown that the model is capturing this weakening of the gyre that is consistent with the decreases in mixing. It is possible that the sparse observations by Corbière et al. (2007) did not capture these DIC decreases. However, it is also possible that the model overestimates them.

Nevertheless, our modeled increase in the CO$_2$ flux in the subpolar gyres (18% for the 50-80N region) is, however, also consistent with the atmospheric CO$_2$ inversion of Rödenbeck (2005) who find a 20% flux increase in the subpolar North Atlantic from 50-80N for the period 1996-2004 (LeQuéré et al., 2007; Rödenbeck et al., 2003). Clearly inconsistencies remain in our understanding of the CO$_2$ sink in this region. Perhaps the observations discussed in Corbiere et al. (2007) in this region are too sparse, spatially and temporally, to offer results that can be extrapolated over the whole gyre. The model does suggest that changes in pCO$_2$ can be highly localized with spatial heterogeneities making it difficult to extrapolate data to nearby regions or across the basin. However, it is also possible that the model is overestimating the changes in deep mixing of the region. Such discrepancies between the in situ observations and the model highlight the need for further surface carbon observations in this region and neighboring areas to help constrain the magnitude and direction of the flux trends in the subpolar gyre.

5. Conclusions

We have used a regional ocean biogeochemical model of moderate complexity to assess the trends in the carbon sink and other related parameters in the North Atlantic from 1992-2006. The model for the most part captures the pCO$_2$ and flux trends observed at BATS and along VOS shipping tracks (Bates, 2007; Schuster and Watson, 2007). This model shows that strong,
regionally distinct trends in the CO$_2$ flux and pCO$_2$ relate to profound change in physical and
chemical state of the basin over the 15-year period.

Schuster and Watson (2007) argue that there has been a reduction in the carbon sink of
~0.24 Pg C yr$^{-1}$ from the mid-1990s to the early-2000s over a broad area from 20N to 65N.
While the results of this model agree with the basis for these findings in all regions except the
western subpolar gyre, we show that any reduction in the sink may be due to a shift in a number
of the mechanisms driving the carbon sink such as SST warming and excess DIC accumulation
due to circulation and mixing changes. Certainly there has been an increase in atmospheric CO$_2$
during this time period, and such an increase will drive more CO$_2$ into the surface ocean, limited
by the buffering capacity of seawater (i.e. the Revelle Factor). The results of this model,
however, show that in the subpolar gyre, surface pCO$_2$ may have increased much more slowly
than the atmosphere. This analysis is highlighted to stress that the ocean carbon cycle is a four
dimensional entity (time included) and that integrating over broad regions, such as in Schuster
and Watson (2007), may be smoothing over spatial heterogeneities in the carbon system, such as
changes due to surface forcing and dynamical transport.

The subpolar gyre exhibits dynamics-driven controls on pCO$_2$, such that changes in
mixing have the greatest impact on the overall pCO$_2$ variability. While the modeled flux trend in
this region does not match the observed trend from sparsely sampled in situ observations
(Corbière et al., 2007), the modeled SST and freshwater changes that drive such changes in
mixing are consistent with hydrographic observations (ICES, 2006; Lazier et al., 2002;
Yashayaev, 2007), suggesting that the model is appropriately capturing some of the major
mechanisms in the subpolar gyre, including circulation changes seen in Häkkinen and Rhines
(2004). Similarly, an atmospheric inversion (LeQuéré et al., 2007; Rödenbeck et al., 2003;
Rödenbeck, 2005) suggests an increasing CO\textsubscript{2} uptake in the subpolar gyre over the analysis period that is consistent with the model. Our analysis of the pCO\textsubscript{2} components and the DIC diagnostics of this region show that the modeled decline in pCO\textsubscript{2} in the subpolar gyre over this period is primarily driven by the decrease in vertical supply of DIC, driven by shoaling of MLDs. The physical forcings controlling mixing appear to be a dominant mechanism of pCO\textsubscript{2} variability, particularly in the western subpolar gyre/Labrador Sea region.

The pCO\textsubscript{2} in the basin south of 45N is strongly temperature driven. The trends in SST, DIC, and pCO\textsubscript{2} are consistent with BATS timeseries data and VOS observations. We show that the spatial variability of the pCO\textsubscript{2} trend in this region is primarily driven by SST, but the positive nature of the overall pCO\textsubscript{2} trend across the entire basin occurs from a pCO\textsubscript{2}-DIC trend that overcompensates the opposing pCO\textsubscript{2}-ALK. DIC diagnostics in this region do not show a clear dominating mechanism. Vertical mixing, biology, freshwater and horizontal transport all contribute to the pCO\textsubscript{2}-DIC trends.

While spatial patterns of pCO\textsubscript{2} change between mid-1990s and mid-2000s appear to be strongly related to variability in climate forcing, the overall magnitude of the trends are driven by the increasing trend in atmospheric CO\textsubscript{2}. Results from a preindustrial simulation show that the spatial nature of the trends in pCO\textsubscript{2} and its components is nearly identical to that of the modern simulation, indicating the patterns are driven by the physical forcing and subsequent changes to ocean circulation and mixing.

Complete timeseries data (i.e. BATS) has been particularly useful in our understanding of the North Atlantic carbon sink, but this data is limited to the subtropical/temperature-driven regions of the basin. Establishment of a time series and/or more observations in the subpolar/dynamics-driven region of the basin would be useful in further assessment of trends in
the North Atlantic. Combined with a need for continued observational data, future modeling
work is also needed. In particular, analysis of longer model runs will help to assess longer
timescale trends and patterns of variability, and also to further elucidate mechanistic
relationships.

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Foundation for funding.
Figure 1. Comparison of (top) $pCO_2$, (middle) SST, and (bottom) DIC between model results and observations at the Bermuda Atlantic Time-series Study (BATS)/Hydrostation S from 1983 through 2006 (Bates, 2007).
Figure 2. Correlation plots. (A) pCO$_2$ and SST, (B) pCO$_2$ and MLD. Correlation coefficients were calculated from fields that were detrended and smoothed over 12 months to remove the seasonal cycle. Dashed contours indicate negative values. Dark solid line indicates zero line. White areas indicate those regions where the correlation coefficient is not significantly different from zero at the 95% level.
Figure 3. (A) Basin averaged CO$_2$ flux for 14-50N and 50-79N for modern (bold) and preindustrial (thin), subpolar (dash) and subtropics (solid). Also included on the plot are 1995 observational data points. These data points use Takahashi et al. (2002) $\Delta p$CO$_2$ to calculate the flux using Wanninkhof (1992) with updated coefficients from Sweeney et al. (2007). (B) Trend in the CO$_2$ Flux, calculated by taking the difference between the 2003-2006 average and the 1992-1995 average. Dashed contours indicate negative values. Dark solid line indicates zero line.
Figure 4. First Empirical Orthogonal Functions of surface (A) pCO$_2$ and (B) CO$_2$ flux. Dashed contours indicate negative values. The dark solid line indicates zero line. (C) First standardized principal components of pCO$_2$ and the CO$_2$ flux. The two PC1s are highly correlated ($r=0.87$), suggesting that much of the variability in the air-sea flux of CO$_2$ is driven by the surface pCO$_2$. 
Figure 5. Trend in the (A) $pCO_2$, (B) $pCO_2$-ALK, (C) $pCO_2$-DIC, (D) $pCO_2$-SST, calculated by taking the difference between the 2003-2006 averages and the 1992-1995 averages. Dashed contours indicate negative values. Areas with values between -3 and 3 µatm have been whited-out to accentuate smaller differences around the zero line (bold contour). Note that (A) has different color bar than (B), (C) and (D).
Figure 6.
Figure 6. Plots of the DIC Diagnostics: (A) DIC-vertical, (B) DIC-biology, (C) DIC-fresh, (D) DIC-horizontal, and (E) DIC-flux. The first column of plots shows the mean of values from 1992-95 for each diagnostic, the second column shows the mean for 2003-06, and the third column shows the difference between the two (2003-06 minus 1992-95). All diagnostics were calculated using a depth-weighted average over the top 100m to account for changes that extend below the uppermost 10m, such as biological productivity. The heavy contour indicates the zero line.
Figure 7. Trend in the (A) Mixed Layer Depths and (B) PO4 concentration, calculated by taking the difference between the 2003-2006 averages and the 1992-1995 averages. Dashed contours indicate negative values. Dark solid line indicates zero line.
Figure 8. Preindustrial run trends in the (A) pCO₂, (B) pCO₂-ALK, (C) pCO₂-DIC, (D) pCO₂-SST, calculated by taking the difference between the 2003-2006 averages and the 1992-1995 averages. Dashed contours indicate negative values. Dark solid line indicates zero line. Note that (A) has different color bar than (B), (C) and (D).
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Figure Captions

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