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2016

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### Recommended Citation

Cahill, Bronwyn; Wilkin, John; Fennel, Katja; Vandemark, Doug; and Friedrichs, Marjorie A.M., "Interannual and seasonal variabilities in air-sea CO<sub>2</sub> fluxes along the US eastern continental shelf and their sensitivity to increasing air temperatures and variable winds" (2016). *VIMS Articles*. 244.

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## RESEARCH ARTICLE

10.1002/2015JG002939

## Special Section:

Carbon and Nitrogen Fluxes at the Land-Ocean Interface

## Key Points:

- Air-sea CO<sub>2</sub> fluxes are simulated using coupled biogeochemical-circulation model
- Shelf air-sea CO<sub>2</sub> fluxes respond differently to variable atmospheric forcing
- Temperature is very important in regulating air-sea CO<sub>2</sub> flux variability

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## Citation:

Cahill, B., J. Wilkin, K. Fennel, D. Vandemark, and M. A. M. Friedrichs (2016), Interannual and seasonal variabilities in air-sea CO<sub>2</sub> fluxes along the U.S. eastern continental shelf and their sensitivity to increasing air temperatures and variable winds, *J. Geophys. Res. Biogeosci.*, 121, 295–311, doi:10.1002/2015JG002939.

Received 30 JAN 2015

Accepted 19 DEC 2015

Accepted article online 28 DEC 2015

Published online 3 FEB 2016

## Interannual and seasonal variabilities in air-sea CO<sub>2</sub> fluxes along the U.S. eastern continental shelf and their sensitivity to increasing air temperatures and variable winds

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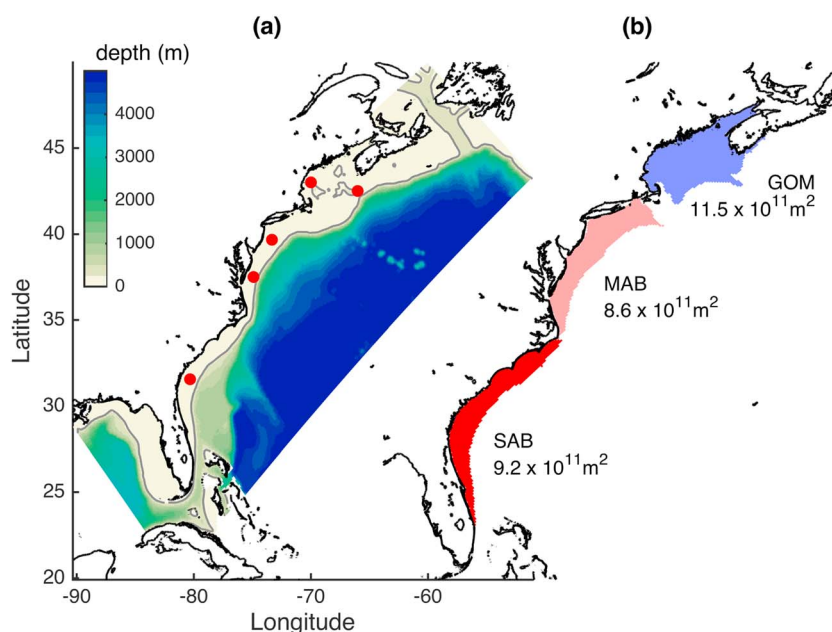
**Abstract** Uncertainty in continental shelf air-sea CO<sub>2</sub> fluxes motivated us to investigate the impact of interannual and seasonal variabilities in atmospheric forcing on the capacity of three shelf regions along the U.S. eastern continental shelf to act as a sink or source of atmospheric CO<sub>2</sub>. Our study uses a coupled biogeochemical-circulation model to simulate scenarios of “present-day” and “future-perturbed” mesoscale forcing variability. Overall, the U.S. eastern continental shelf acts as a sink for atmospheric CO<sub>2</sub>. There is a clear gradient in air-sea CO<sub>2</sub> flux along the shelf region, with estimates ranging from  $-0.6 \text{ Mt C yr}^{-1}$  in the South Atlantic Bight (SAB) to  $-1.0 \text{ Mt C yr}^{-1}$  in the Mid-Atlantic Bight (MAB) and  $-2.5 \text{ Mt C yr}^{-1}$  in the Gulf of Maine (GOM). These fluxes are associated with considerable interannual variability, with the largest interannual signal exhibited in the Gulf of Maine. Seasonal variability in the fluxes is also evident, with autumn and winter being the strongest CO<sub>2</sub> sink periods and summer months exhibiting some outgassing. In our future-perturbed scenario spatial differences tend to cancel each other out when the fluxes are integrated over the MAB and GOM, resulting in only minor differences between future-perturbed and present-day air-sea CO<sub>2</sub> fluxes. This is not the case in the SAB where the position of the along-shelf gradient shifts northward and the SAB becomes a source of CO<sub>2</sub> to the atmosphere ( $0.7 \text{ Mt C yr}^{-1}$ ) primarily in response to surface warming. Our results highlight the importance of temperature in regulating air-sea CO<sub>2</sub> flux variability.

### 1. Introduction

The flux of CO<sub>2</sub> between the atmosphere and the ocean is a critical process in the global carbon cycle, yet significant uncertainty in estimates of this flux stem from the portion associated with uptake and release of CO<sub>2</sub> on continental shelves. Globally, it is estimated that approximately 26% (or  $2.3 \pm 0.4 \text{ Pg C yr}^{-1}$ ) of CO<sub>2</sub> presently emitted as a result of anthropogenic activities is absorbed by the world ocean [Le Quéré *et al.*, 2009, 2010]. In the open ocean, Takahashi *et al.* [2009] estimate a contemporary sink of atmospheric CO<sub>2</sub> of  $-1.6 \pm 0.9 \text{ Pg C yr}^{-1}$ . In the coastal ocean, Laruelle *et al.* [2014] have recently evaluated the flux of CO<sub>2</sub> as a sink of atmospheric CO<sub>2</sub> of  $-0.19 \pm 0.05 \text{ Pg C yr}^{-1}$ . While this corresponds to 8% (or 14%) of the open ocean sink, its significance is clouded by major uncertainties associated with the estimates. Regional quantification and global extrapolation of coastal fluxes are uncertain due to spatial and temporal variabilities and undersampling [Fennel, 2010].

High productivity on shelf systems is driven by the interplay between temperature, light, and nutrient inputs from land, remineralization in shelf sediments due to tight benthic-pelagic coupling that returns nutrients to the euphotic zone on time scales on the order of a year and most importantly by upwelling and onwelling of nutrients from the open ocean. However, not all continental shelf systems respond in the same manner to these drivers and there are wide regional differences in continental shelves' potential to be a source or sink for atmospheric CO<sub>2</sub> [Cai *et al.*, 2006; Borges, 2011; Borges *et al.*, 2005]. This makes it important to view regions as distinct provinces.

Evaluating and understanding air-sea CO<sub>2</sub> fluxes on the scale of continental shelves is difficult; we adopt an approach that combines model experiments with the analysis of in situ data and considers ocean circulation, primary production, and geochemistry. A key factor influencing variability of air-sea CO<sub>2</sub> fluxes in shelf regions is atmospheric forcing. In this study, we investigate the impact of interannual and seasonal variabilities in atmospheric forcing on the capacity of different shelf subregions to act as a sink or source of atmospheric CO<sub>2</sub>. We



**Figure 1.** (a) Study location, model domain, and location of observations (red circles) used in the analyses (black contour denotes the location of 200 m isobath) and (b) boundaries and surface area of the analysis regions, Gulf of Maine (GOM), Mid-Atlantic Bight (MAB), and South Atlantic Bight (SAB).

assess the “present-day” response of different shelf subregions to contemporary atmospheric forcing and evaluate the sensitivity of regional air-sea  $\text{CO}_2$  fluxes to a “future-perturbed” scenario where the present-day atmospheric forcing is perturbed with atmospheric anomalies derived from modern and future scenarios of a regional climate model (RegCM3). We focus only on changes in atmospheric forcing and do not take into account changes in river inputs, projected future ocean biogeochemical state, or higher atmospheric  $p\text{CO}_2$  ( $p\text{CO}_2^{\text{AIR}}$ ). We assume, to first order, that the rate of change in  $p\text{CO}_2^{\text{SEA}}$  in the coastal ocean is tracking the rate of  $p\text{CO}_2^{\text{AIR}}$  increase, so that the air-sea gradient of  $\text{CO}_2$  does not change over time, at least for surface waters, between our present-day and future-perturbed scenarios. We provide further details in section 3.2 explaining the reasons justifying this assumption.

The rest of the paper is organized as follows: in section 2, we present the overall study region; the Northeast North American shelf; and the different characteristics of three subregions under investigation: Gulf of Maine, Mid-Atlantic Bight, and South Atlantic Bight. In section 3, we present the biogeochemical model, the configuration of our model experiments, and the approach to our sensitivity analysis. In the first part of section 4, we present the model results and an assessment of present-day seasonal and interannual air-sea  $\text{CO}_2$  fluxes for the three subregions, including an evaluation of the modeled fluxes in comparison to observations. In the second part of section 4, we present the results of a sensitivity analysis and the subregional responses to future-perturbed variations in atmospheric forcing. A discussion and conclusions are presented in section 5.

## 2. Study Location

The area of interest is the Northeast North American (NENA) continental shelf region along the eastern seaboard of the United States (Figure 1a). Three subregions have been selected for detailed analysis (Figure 1b). These are the Gulf of Maine (GOM), the Mid-Atlantic Bight (MAB), and the South Atlantic Bight (SAB). The choice of boundaries circumscribing each of these regions is broadly determined by the differences in their physical and biological regimes and the 200 m bathymetric contour. They are consistent with those used in *Hofmann et al.* [2011] and *Fennel* [2010]. Primary productivity is high in all three regions, but they have distinct characteristics that make them suitable candidates for evaluating the response of air-sea  $\text{CO}_2$  fluxes to different regimes.

### 2.1. Gulf of Maine

The Gulf of Maine (GOM) is a marginal sea at temperate latitudes in the Northwest Atlantic bounded by Cape Cod to the south and Nova Scotia to the east and separated from the open ocean by Georges Bank and

Browns Bank. With the exception of the Boston region, the coastal watershed is sparsely populated and distinctively rural along much of the coastline. There is a seasonal cycle in stratification and significant primary productivity ( $>270 \text{ g cm}^{-2} \text{ yr}^{-1}$ ) [O'Reilly and Busch, 1984; Balch et al., 2008]. Seasonal and interannual circulation variabilities are attributed to variations in shelf-sea exchange through the narrow Northeast Channel and fresher coastal source waters from along the Scotian Shelf [Townsend, 1991; Townsend et al., 2006; Pringle, 2006]. Physical ocean circulation characteristics include a large semidiurnal tide, a persistent counterclockwise gyre circulation with several distinct coastal currents [Pettigrew et al., 2005], uneven coastline and bathymetry, and large seasonal freshwater inflow [Salisbury et al., 2008]. The western Gulf of Maine is seasonally stratified with heat and buoyancy fluxes exceeding tidal and wind-driven mixing forces from roughly March to November each year. The region is also regularly impacted by local and distant river runoff from April to July, with a resulting increase in buoyancy flux and coastal current velocities [Geyer et al., 2004; Salisbury et al., 2008] and decrease in surface water residence times [Manning et al., 2009]. There is significant carbon input to the GOM from rivers, and temperature, tidal, and wind dynamics all conspire to regulate air-sea  $\text{CO}_2$  fluxes in the region [Vandemark et al., 2011].

## 2.2. Mid-Atlantic Bight

The Mid-Atlantic Bight (MAB) is a highly productive ( $>310 \text{ g cm}^{-2} \text{ yr}^{-1}$ ) [O'Reilly et al., 1987] broad continental shelf characterized by consistently high-chlorophyll biomass ( $>1 \text{ mg chlorophyll m}^{-3}$ ) which supports a diverse food web that includes abundant fin and shellfish populations [Yoder et al., 2001]. It extends from Cape Cod, Massachusetts, southward to Cape Hatteras, North Carolina, and extends offshore for several hundreds of kilometers. It drains much of the northeast and Mid-Atlantic U.S. including three major watersheds that flow into the Chesapeake Bay, Delaware Bay, and Hudson Estuary, all of which ultimately discharge freshwater, nutrients and organic carbon to the MAB. It is a densely populated, highly urbanized region. Chesapeake Bay is the largest and most productive estuary in the U.S. Its drainage basin extends from New York to Virginia ( $171,990 \text{ km}^2$ ;  $\sim 60\%$  forested), and it discharges more freshwater (mean annual discharge of  $2280 \text{ m}^3 \text{ s}^{-1}$  [Schubel and Pritchard, 1986]) than any other river/estuary system along the U.S. Atlantic coast, contributing about half the freshwater that flows into the MAB. The discharge from the Hudson-Raritan estuary system adjacent to New York City accounts for  $\sim 20\%$  of the freshwater received by the MAB [Taylor et al., 2003], while the urbanized Delaware Estuary has a mean annual freshwater discharge of  $550 \text{ m}^3 \text{ s}^{-1}$  [Lebo and Sharp, 1993], contributing  $\sim 15\text{--}20\%$  of the freshwater discharge entering the MAB.

The waters of the MAB exhibit pronounced seasonal and interannual variabilities in temperature and salinity [Mountain, 2003]. In late spring and early summer, a strong thermocline (water temperatures can span from  $30$  to  $8^\circ\text{C}$  in  $<5 \text{ m}$ ) develops at about the  $20 \text{ m}$  isobath across the entire shelf, isolating a continuous midshelf "cold pool" (formed in winter months) that extends from Nantucket to Cape Hatteras [Houghton et al., 1982; Biscaye et al., 1994]. The cold pool persists throughout the summer until fall when the water column overturns and mixes [Houghton et al., 1982]. Thermal stratification redevelops in spring as the frequency of winter storms decrease and surface heat flux increases [Lentz et al., 2003].

## 2.3. South Atlantic Bight

The South Atlantic Bight (SAB) extends along the southeastern United States coast from West Palm Beach, Florida, to Cape Hatteras, North Carolina. It is characterized by a narrow, shallow shelf with the Gulf Stream flowing northward in close proximity to the shelf break. A series of barrier islands delimit the shoreline with extensive salt marshes. The inner shelf ( $<20 \text{ m}$ ) is typically influenced by discharge from rivers and marshes. There are 11 rivers that input approximately  $66 \text{ km}^3$  freshwater annually into the South Atlantic Bight [Menzel, 1993]. River discharge usually peaks between February and April and is at a minimum between September and November. The release of groundwater from salt marshes also contributes to the freshwater input into the region, although this flux is poorly quantified. The total input of organic carbon from marshes and rivers in the South Atlantic Bight is the highest along the U.S. East Coast [Hopkinson, 1985; Wang and Cai, 2004]. The middle ( $20\text{--}40 \text{ m}$ ) and outer shelves ( $40\text{--}60 \text{ m}$ ) are influenced less by river discharge and more by nutrient-rich water intrusions occurring within a semipermanent gyre that exists downstream of the bathymetric feature called the "Charleston Bump" off Long Bay, South Carolina. This brings nutrient-rich water from off the shelf edge and results in enhanced biological production [Bane and Dewar, 1988]. Annual primary production estimates are given as  $320 \text{ g cm}^{-2} \text{ yr}^{-1}$  [Menzel, 1993].

The annual temperature cycle in the SAB exhibits important seasonal characteristics. Sea surface temperature is relatively uniform over the whole shelf in summer, between 28°C and 31°C in July; however, large cross-shelf gradients in sea surface temperature persist in the winter, ranging from 10°C on the inner shelf to 25°C on the outer shelf [Jiang *et al.*, 2008].

### 3. Model

#### 3.1. NENA Model

The NENA model was developed to investigate the transport and cycling of carbon and nitrogen to and within the U.S. East Coast coastal ocean margin and the impact of climate variability, climate change, and land cover/land use change on these fluxes [Fennel *et al.*, 2006; Hofmann *et al.*, 2008, 2011]. NENA is based on the Regional Ocean Modeling System (ROMS) and a coupled biogeochemical module. ROMS is widely used for shelf circulation and coupled physical-biological applications [e.g., Dinniman *et al.*, 2003; Lutjeharms *et al.*, 2003; Marchesiello *et al.*, 2003; Peliz *et al.*, 2003; Fennel *et al.*, 2006, 2008; Wilkin, 2006]. The ROMS computational kernel [Shchepetkin and McWilliams, 1998, 2003, 2005] produces accurate, conservative evolution of tracer fields, which is a particularly attractive feature for biogeochemical modeling because it facilitates accurate interaction among tracers and accounting of total nutrient and carbon budgets.

The NENA model domain (Figure 1a) encompasses the entire U.S. East Coast continental shelf, which presents a number of challenges in terms of model complexity, boundaries, and resolution. There are 31 rivers represented along the land-ocean boundary, complex open boundaries that include subtropical and subpolar regions of the Atlantic Ocean, and a significant shelf area where sediment-water interactions play an important role [e.g., Fennel *et al.*, 2006]. The model configuration used here has a 10 km horizontal resolution and 30 terrain-following vertical levels stretched to give increased resolution in surface and bottom boundary layers. This is sufficient to capture the dominant dynamics governing shelf-wide circulation. Open boundary temperature, salinity, and subtidal frequency velocity are taken from 5 day averages of the HYbrid Coordinate Ocean Model (HYCOM) data assimilation product developed as part of the Global Ocean Data Assimilation Experiment North Atlantic Basin “best estimate” analysis for 2003 to the present [Chassignet *et al.*, 2007]. Tides are introduced at the boundary using harmonic data from the Oregon State University TOPEX/Jason altimeter data inversion [Egbert and Erofeeva, 2002] and a surface gravity wave radiation scheme [Flather, 1976]. Air-sea heat and momentum fluxes are computed using bulk formulae [Fairall *et al.*, 2003] applied to model sea surface conditions and air temperature, pressure, humidity, radiation, precipitation, and winds from 3 h interval National Center for Environmental Prediction North American Regional Reanalysis (NCEP-NARR) [Mesinger *et al.*, 2006]. Vertical turbulent mixing closure uses the parameterization of Mellor and Yamada [1982] implemented via the *k-kl* option of Warner *et al.* [2005].

The biogeochemical module of the NENA model incorporates carbon and nitrogen dynamics after Fennel *et al.* [2006, 2008] and Fennel and Wilkin [2009]. The model currency is nitrogen with an assumed fixed C:N ratio for plankton but independent C and N state variables for detritus. Particulate flux to the seabed is partly recycled and partly lost to N<sub>2</sub> gas via an explicit sediment denitrification parameterization. The Ocean Carbon Cycle Model Intercomparison Project standard is used for the carbonate system and the Wanninkhof [1992] formulation for gas exchange. Atmospheric  $p\text{CO}_2^{\text{AIR}}$  is fixed at 377.4 ppm. Boundary estimates for nitrate, dissolved inorganic carbon (DIC), total alkalinity (TA), and oxygen were derived using temperature and salinity data from the World Ocean Atlas [Conkright *et al.*, 2002] and property-property relationships, which account for regional differences in the subpolar and subtropical regions based on Fennel *et al.* [2008], Lee *et al.* [2000], and Millero *et al.* [1998]. Inputs from 31 rivers are prescribed as a point source climatology derived from U.S. Geological Survey freshwater gauge data after Howarth *et al.* [2000], essentially representing net freshwater and biogeochemical (total nitrogen in the nitrate pool) transport into the coastal ocean. There is no interannual variability in our river inputs.

#### 3.2. Model Experiments

Two model scenarios were developed and run for a 4 year period corresponding to 2004 to 2007 (Table 1). The first scenario, referred to as present day, represents contemporary mesoscale variability in forcing as captured by NARR-NCEP 3 hourly meteorology fields from 2004 to 2007. The year 2004 corresponds to a positive North Atlantic Oscillation (NAO) index, while 2005 to 2007 corresponds with a negative NAO index. The difference between the positive and negative NAO periods is evident in the wind data, where the Gulf of Maine

**Table 1.** Configuration of “Present-Day” and “Future-Perturbed” Scenarios

	Present Day	Future Perturbed
Atmospheric forcing	NCEP-NARR 3 h fields • $T_{AIR}$ • $P_{AIR}$ • $Q_{AIR}$ •RAIN •SWRAD •LWRAD • $U_{WIND}$ • $V_{WIND}$	Atmospheric anomalies were added to present-day NCEP-NARR fields. Anomalies were derived from two 10 year simulations of RegCM3 model [Chen <i>et al.</i> , 2003] representing present and end of century (doubled) CO <sub>2</sub> levels, forced by 100 year transient run of NCAR climate system model
$pCO_2^{AIR}$		377.4 ppmv
Time period	4 years corresponding to 2004 to 2007	

and Mid-Atlantic Bight experience pronounced northeasterly winds in 2004, as compared to pronounced easterly winds in these regions in subsequent years (not shown).

The second scenario, referred to as future perturbed, adjusts the present-day forcing by adding atmospheric forcing anomalies derived from modern and future scenarios of a regional climate model, RegCM3, indicative of a doubling of atmospheric CO<sub>2</sub>. River inputs remain

the same as in the present-day scenario. We intentionally do not have any future-perturbed signal in the river inputs because we want to isolate the impact of atmospheric variability on air-sea CO<sub>2</sub> fluxes.

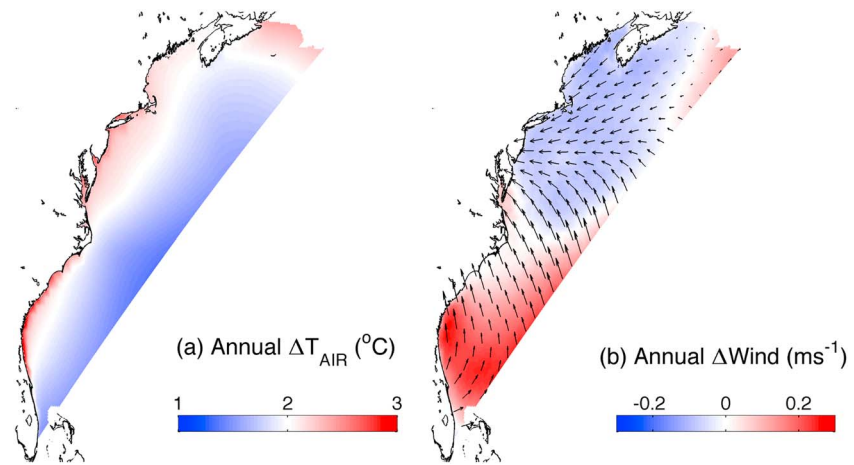
The concentration  $pCO_2^{AIR}$  was kept at 377.4 ppm, as in the present-day scenario for several important reasons. We do not take into account the projected biogeochemical ocean state because of the absence of robust, regional future projections of ocean state required to initialize a future air-sea biogeochemical equilibrium. We note that Earth system models (ESMs) which have run coupled biophysical projections of future ocean state (i.e., various Intergovernmental Panel on Climate Change (IPCC) scenarios) are typically global simulations where the biogeochemical equilibrium is maintained throughout the duration of the simulation (from present to future). In other words, the biogeochemical ocean state evolves in concert with increasing atmospheric CO<sub>2</sub> concentrations and is not abruptly imposed in a regional context, at some point in the future, as would be the case if we were to double the concentration of  $pCO_2^{AIR}$  in our future-perturbed scenario. Moreover, global ESM simulations do not capture the open ocean mesoscale dynamics or coastal boundary current inflows from the north (Labrador) and south (Gulf of Mexico) that we need to use in our regional model context. A recent comparison of selected physical variables for the NW Atlantic from historical and future simulations (IPCC RCP4.5 and RCP8.5) from six ESMs indicates that all six models have a poor representation of the detailed structure of important ocean and ice features. Furthermore, it is doubtful whether any of the models have an adequate representation of regional ocean dynamics to reliably simulate ocean climate variability and change. The magnitudes of the intermodel differences in the projected changes are comparable to those of the ensemble mean changes, such that it is not possible to make any robust quantitative projections at a regional scale, yet it is unclear whether any of the models examined are worse than the others to the point that they should be rejected from an ensemble approach to climate change projections [Loder and van der Baaren, 2013]. Instead, we assume to first order that the rate of change in  $pCO_2^{SEA}$  in the coastal ocean is tracking the rate of  $pCO_2^{AIR}$  increase, so that the air-sea gradient of CO<sub>2</sub> does not change over time, at least for surface waters, which is consistent with the space and time scales of our study.

As an added check, we looked into the potential impact on air-sea flux from a Revelle factor change due to an order of +100 ppmv change in atmospheric CO<sub>2</sub> in this region. We find a small increase in the surface Revelle factor in the SAB (approximately 2%) and even less in the MAB and GOM regions ( $\pm 0.5\%$ ). While this could impact solubility by a small amount from one scenario to another (e.g., due to changes in temperature), the net impact on our study conclusions appears to be quite small.

The main features of the atmospheric anomalies imposed in the future-perturbed scenario are found in the air temperature and wind anomalies. There is an overall increase in air temperature over the entire model domain ranging from 1°C offshore to as much as 3°C along coastal regions. This will have important consequences for the modeled surface temperature and  $pCO_2^{SEA}$ . There is also a decrease in the northeasterly wind component in the Gulf of Maine and northern Mid-Atlantic Bight, an increase in the easterly wind component in the southern Mid-Atlantic Bight, and an increase in the southeasterly wind component in the South Atlantic Bight (Figure 2).

### 3.3. Sensitivity Analysis

The modeled local rate of change of DIC accounts for biogeochemical sources and sinks, advection and diffusion by ocean circulation, and the exchange of CO<sub>2</sub> gas across the air-sea interface. Schematically, this modeled rate may be described as



**Figure 2.** RegCM3-derived (a) annual air temperature and (b) wind speed atmospheric anomalies.

$$\begin{aligned} \partial \text{DIC} / \partial t = & - \text{nutrient-based uptake by phytoplankton} \\ & + \text{excretion (due to basal metabolism and grazing dependence)} \\ & + \text{solubilization of detritus small and large} \\ & - \text{advection} + \text{diffusion} + \text{air-sea flux of CO}_2 \end{aligned}$$

Details on the parameterization of the biogeochemical processes are given by *Fennel et al.* [2006, 2008] and *Druon et al.* [2010].

The air-sea flux of CO<sub>2</sub> is parameterized by

$$F = \frac{kw^2}{Sc^{1/2}\Delta z} \text{CO}_2^{\text{SOL}} (p\text{CO}_2^{\text{AIR}} - p\text{CO}_2^{\text{SEA}}) \quad (1)$$

which is a function of wind speed,  $w$ ; the Schmidt number ( $Sc$ ) for CO<sub>2</sub> which depends on temperature,  $T$ ; the solubility of CO<sub>2</sub> (a function of  $T$  and  $S$ ); and the air-sea CO<sub>2</sub> partial pressure difference (a function of  $T$ ,  $S$ ,  $T_A$ , and  $\text{DIC}$ ).  $\Delta z$  represents the thickness of the top model grid box. In order to explore the sensitivity of air-sea CO<sub>2</sub> fluxes to variability in atmospheric forcing, we examine the contributions of these four terms to the change in air-sea CO<sub>2</sub> flux between our “present-day” and “future-perturbed” scenarios. We note that our choice of the *Wanninkhof* [1992] gas transfer parameterization,  $k$ , predates more recent developments and improvements in this field [*Wanninkhof*, 2014]. The uncertainty associated with the choice of gas transfer velocity formulation is estimated to be around 10% for wind speeds between 3 and 12 ms<sup>-1</sup> [*Ho et al.*, 2011; *Sweeney et al.*, 2007; *Nightingale et al.*, 2000a, 2000b] and as much as 20% for wind speeds > 12 ms<sup>-1</sup> [*Wanninkhof*, 2014].

Following the methodology of *Previdi et al.* [2009], we express the difference  $\delta F$  in the CO<sub>2</sub> flux using first- and second-order Taylor series approximations. We estimate the linear sensitivity of  $F$  to changes in forcing from first-order partial derivatives (equation (2)) and the nonlinear effects contributing to  $\delta F$  by computing second-order partial derivatives (equation (3)).

The first-order partial derivatives are approximated using a method of single field substitution:

$$\frac{\partial F}{\partial X_i} \delta X_i \approx F\left(\underline{X}^{\text{FUTURE}} + \underline{e}_i \Delta X\right) - F\left(\underline{X}^{\text{FUTURE}}\right) \quad (2)$$

where the first term on the right-hand side is calculated using the time series of variable  $X_i$  from the present-day scenario in the term  $\Delta X$  and the time series of all other variables from the future-perturbed scenario in the term  $\underline{X}^{\text{FUTURE}}$ . In the second term on the right-hand side of equation (2), all variables in the term  $\underline{X}^{\text{FUTURE}}$  are from the future-perturbed model scenario. This approach was introduced in meteorological studies of cloud radiative feedback by *Wetherald and Manabe* [1988] and *Colman et al.* [1997]. It provides an estimate of the linear sensitivity of  $F$  to various changes in the system; here the change in  $F$  is due to a change in wind speed, Schmidt number, CO<sub>2</sub> solubility, or  $p\text{CO}_2^{\text{SEA}}$  alone.

Nonlinear effects contributing to  $\delta F$  are estimated by computing second-order partial derivatives. These higher-order derivatives are approximated as

$$\begin{aligned} \frac{\partial^2 F}{\partial X_i \partial X_j} \delta X_i \delta X_j &\approx \left( F \left( X^{\text{FUTURE}} + (e_i + e_j) \Delta X \right) - F \left( X^{\text{FUTURE}} + e_j \Delta X \right) \right) \\ &\quad - \left( F \left( X^{\text{FUTURE}} + e_i \Delta X \right) - F \left( X^{\text{FUTURE}} \right) \right) \\ &= \left( F \left( X^{\text{FUTURE}} + (e_i + e_j) \Delta X \right) - F \left( X^{\text{FUTURE}} + e_j \Delta X \right) \right) - \frac{\partial F}{\partial X_i} \delta X_i \end{aligned} \quad (3)$$

where, again using single field substitution,  $F(X^{\text{FUTURE}} + (e_i + e_j) \Delta X)$  was calculated using the time series of  $X_i$  and  $X_j$  from the present-day scenario, in the term  $\Delta X_i$  and the time series of all other variables from the future-perturbed scenario, in the term  $X^{\text{FUTURE}}$ . The values calculated in equation (3) indicate how the sensitivity of  $F$  to changes in  $X_i$  depends on  $X_j$  or, more generally, how this sensitivity depends on the state of the system. This becomes important in the analysis of the  $p\text{CO}_2^{\text{SEA}}$  contribution to the air-sea  $\text{CO}_2$  flux and how the effects of the different processes which control  $p\text{CO}_2^{\text{SEA}}$  (e.g., net ecosystem production and air-sea gas exchange) depend on the state of the system.

## 4. Results

Previous work on the evaluation of the NENA model supports our confidence in the current estimates of ocean state, including the biogeochemical ocean state [Fennel *et al.*, 2006; Hofmann *et al.*, 2008; Previdi *et al.*, 2009]. Here we focus our evaluation on the modeled air-sea  $\text{CO}_2$  fluxes and their subsequent response to perturbations in atmospheric forcing. To clarify the direction of our air-sea  $\text{CO}_2$  fluxes, a positive flux indicates that the ocean is acting as a source of  $\text{CO}_2$  to the atmosphere, while a negative flux indicates that the ocean is acting as a sink of  $\text{CO}_2$  from the atmosphere.

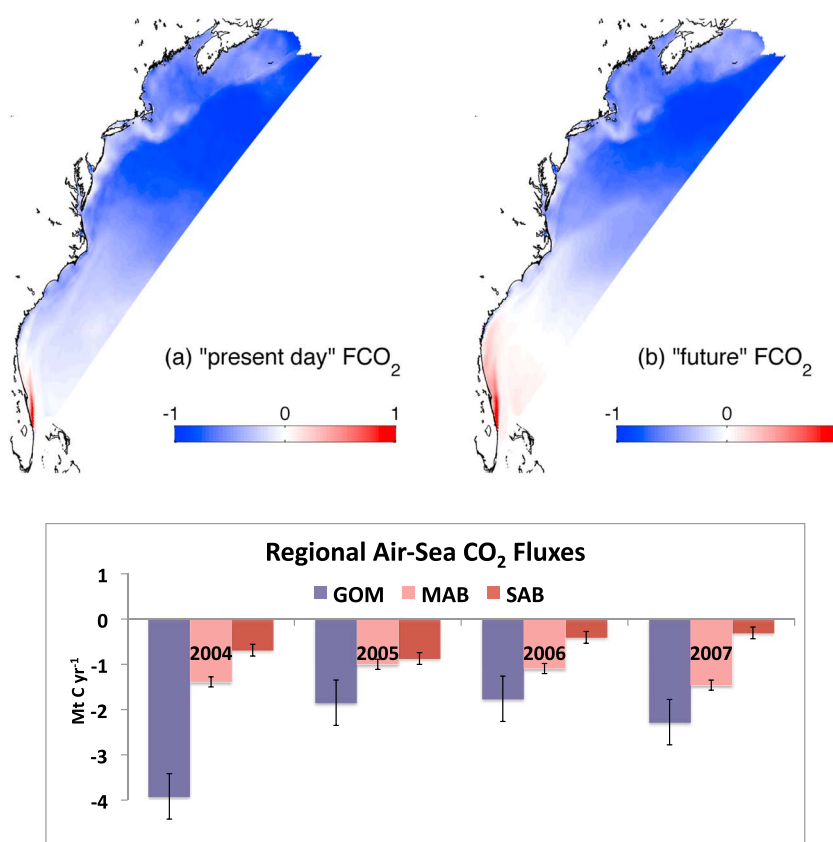
### 4.1. Present Day Air-Sea $\text{CO}_2$ Fluxes

The simulated present-day estimates of air-sea  $\text{CO}_2$  fluxes indicate that on an annual time scale, the Gulf of Maine, Mid-Atlantic Bight, and South Atlantic Bight all act as sinks for atmospheric  $\text{CO}_2$ , with considerable seasonal and spatial variabilities. A clear along-shelf gradient (south to north) in the sign and magnitude of the air-sea  $\text{CO}_2$  flux exists throughout the whole NENA shelf region (Figure 3a), consistent with previous descriptions [Chavez and Takahashi, 2007]. A cross-shelf gradient also exists in the sign and magnitude of the air-sea  $\text{CO}_2$  flux in the South Atlantic and Mid-Atlantic Bights. The inner shelf of the southern part of the South Atlantic Bight acts as a source of  $\text{CO}_2$  to the atmosphere, while the middle and outer shelves in the southern SAB act as a mild sink for atmospheric  $\text{CO}_2$ . This is consistent with the findings of Jiang *et al.* [2008] who report the inner shelf as a source of  $+1.2 \text{ mol C m}^{-2} \text{ yr}^{-1}$  and the middle and outer shelves as sinks of  $-1.23$  and  $-1.37 \text{ mol C m}^{-2} \text{ yr}^{-1}$ , respectively. The northern part of the South Atlantic Bight, however, does not reflect this cross-shelf pattern but shows the inner shelf acting as a mild sink of  $\text{CO}_2$  from the atmosphere and the middle and outer shelves as more or less neutral. Most of the river runoff in the SAB is distributed between  $31^\circ\text{N}$  and  $33^\circ\text{N}$ , with the southern portion of the SAB receiving the least water from rivers [Jiang *et al.*, 2008]. Most likely, our result indicates that we are not resolving the input of organic carbon from marshes and rivers sufficiently in the model in the northern part of the South Atlantic Bight.

The cross-shelf gradient in the Mid-Atlantic Bight is much more consistent with observations. Coastal areas of the Mid-Atlantic Bight act as a source of  $\text{CO}_2$  for the atmosphere, while the Mid-Atlantic Bight shelf and slope waters and the Gulf of Maine act as stronger sinks of atmospheric  $\text{CO}_2$ . A larger interannual variability in air-sea  $\text{CO}_2$  fluxes is evident between 2004 and the subsequent model years (2005–2007), especially in the Gulf of Maine (Figure 3c), possibly in response to NAO-driven variability in atmospheric forcing between 2004 (positive NAO index) and 2005 to 2007 (negative NAO index).

The annual mean estimates of air-sea  $\text{CO}_2$  fluxes show a seasonal cycle within each of the subregions supported by the seasonal cycles of surface temperature,  $p\text{CO}_2^{\text{SEA}}$ , and primary production (Figures 4–6). Autumn and winter are the strongest  $\text{CO}_2$  sink periods, corresponding with lower  $p\text{CO}_2^{\text{SEA}}$  values driven by lower surface

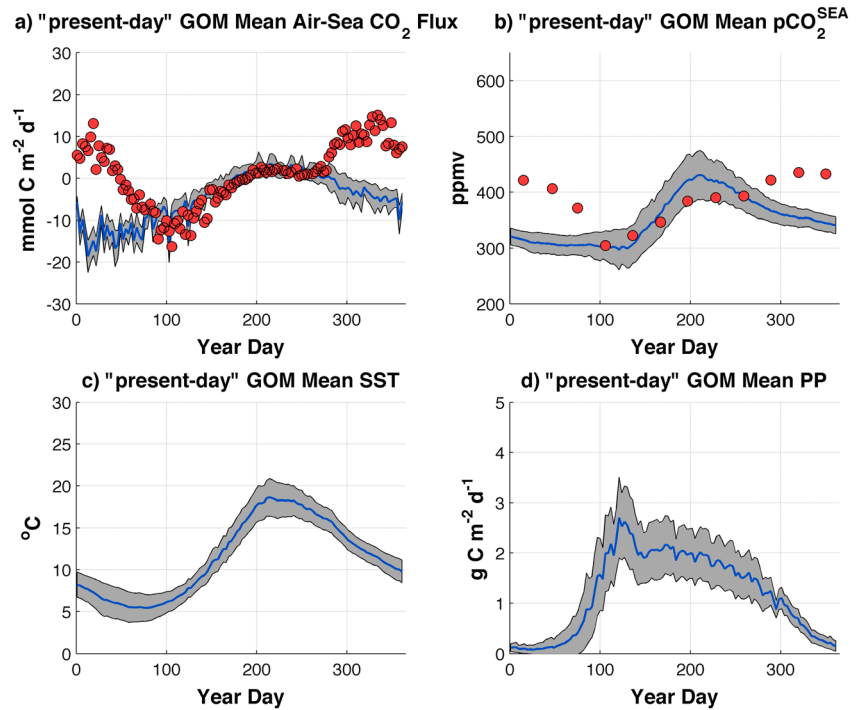




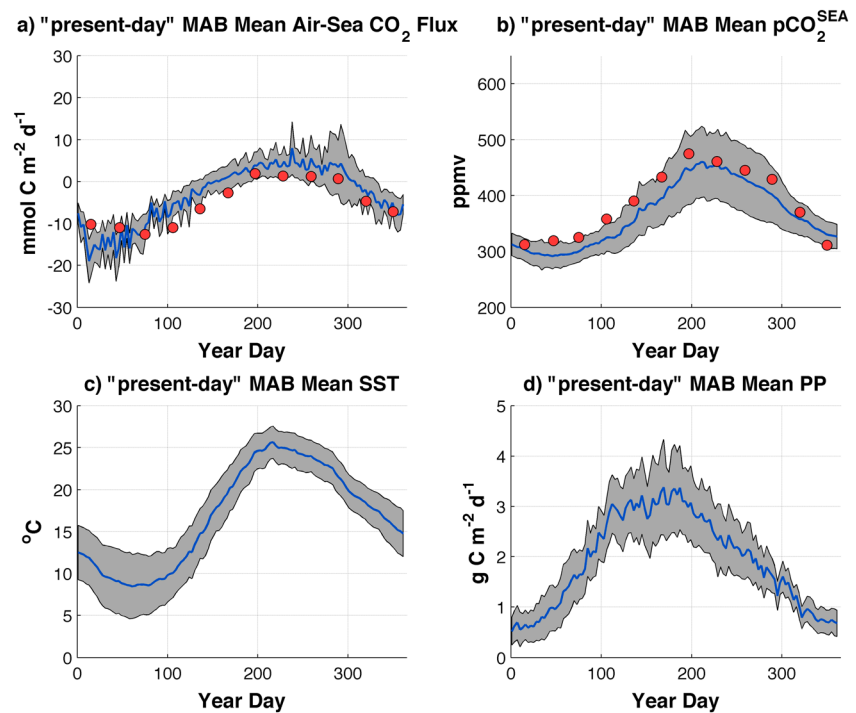
**Figure 3.** Four-year mean (a) "present-day" and (b) "future-perturbed" air-sea CO<sub>2</sub> flux, FCO<sub>2</sub> (mol C m<sup>-2</sup> yr<sup>-1</sup>). (c) Interannual air-sea CO<sub>2</sub> flux, FCO<sub>2</sub> (Mt C yr<sup>-1</sup>).

temperatures. Some outgassing occur during the summer months (June to August) when surface warming tends to increase  $p\text{CO}_2^{\text{SEA}}$ , thus decreasing the uptake of atmospheric CO<sub>2</sub>.

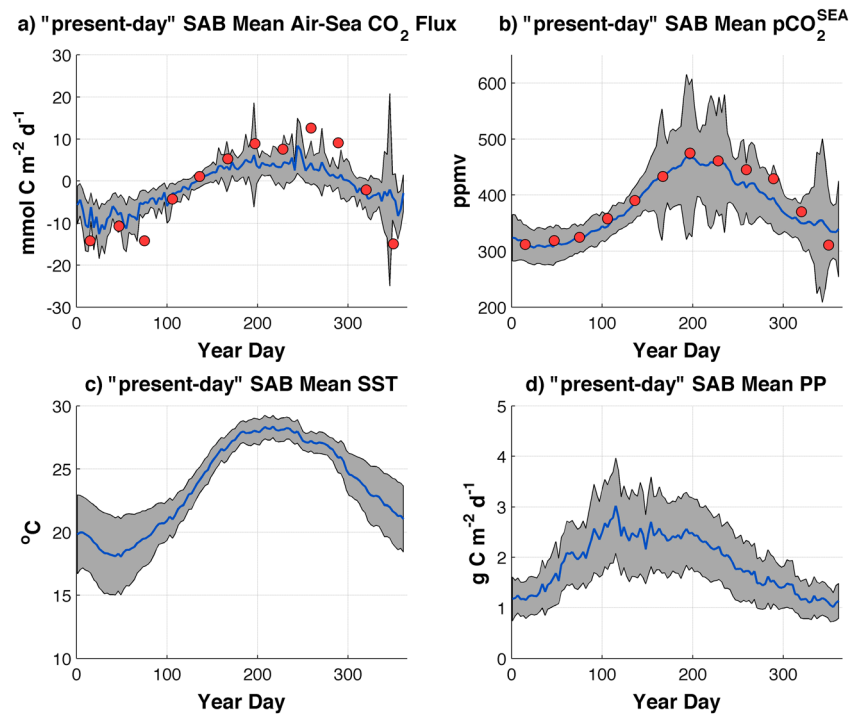
The present-day estimates of annual air-sea CO<sub>2</sub> fluxes agree well with observed and calculated estimates within the MAB and SAB subregions [Takahashi *et al.*, 2009; Jiang *et al.*, 2008] (Table 2). This is also supported by good agreement between the modeled and observed mean  $p\text{CO}_2^{\text{SEA}}$  (Figures 5a and 5b and 6a and 6b). Various other estimates of air-sea CO<sub>2</sub> fluxes for the MAB (−0.6 to −1.7 mol C m<sup>-2</sup> yr<sup>-1</sup>) [Signorini *et al.*, 2013; Fennel *et al.*, 2008; Previdi *et al.*, 2009; DeGrandpre *et al.*, 2002] and SAB (−0.79 mol C m<sup>-2</sup> yr<sup>-1</sup>) [Signorini *et al.*, 2013] regions are also consistent with our modeled estimates. The model seems to capture both the magnitude of the annual air-sea CO<sub>2</sub> flux and also its seasonal cycle for both the Mid-Atlantic Bight and the South Atlantic Bight. This is not the case for the Gulf of Maine, where the annually integrated model results indicate that the region acts as a sink for CO<sub>2</sub> (−1.8 mol C m<sup>-2</sup> yr<sup>-1</sup>). Observations from a coastal station coincident with the model run period (2004 to 2008) indicate that the region acts as a weak source of CO<sub>2</sub> to the atmosphere (0.34 mol C m<sup>-2</sup> yr<sup>-1</sup>) [Vandemark *et al.*, 2011]. Signorini *et al.* [2013] also find the Gulf of Maine to be a weak source of CO<sub>2</sub> to the atmosphere (0.11 mol C m<sup>-2</sup> yr<sup>-1</sup>). While the model captures the observed summer time air-sea CO<sub>2</sub> flux in the Gulf of Maine very well with respect to Vandemark *et al.* [2011], there is a significant divergence in the flux estimates between the model and observations in autumn and winter where the observations show this to be a period of efflux in the region. This is also reflected between the modeled and observed estimates of  $p\text{CO}_2^{\text{SEA}}$  (Figures 4a and 4b). Spatial variations in  $p\text{CO}_2^{\text{SEA}}$  and air-sea CO<sub>2</sub> fluxes will be greater in the 30–45 day spring and fall periods when the mixed layer is in transition [Vandemark *et al.*, 2011; Salisbury *et al.*, 2009]. Precipitation and freshwater discharge in spring can lead to freshening and depletion of DIC in surface waters. It is our supposition that the model is not capturing important changes in water mass properties at these transition periods because our initialization of DIC and alkalinity in the Gulf of Maine does not include sufficient samples from the Gulf of Maine. The DIC and TA initial conditions were derived



**Figure 4.** "Present-day" mean (a) air-sea  $\text{CO}_2$  fluxes ( $\text{mmol C m}^{-2} \text{d}^{-1}$ ), (b)  $p\text{CO}_2^{\text{SEA}}$  (ppmv), (c) SST ( $^{\circ}\text{C}$ ), and (d) primary production ( $\text{g C m}^{-2} \text{d}^{-1}$ ) in the Gulf of Maine (shaded area indicates the standard deviation, and red dots indicate the observations from Vandemark *et al.* [2011]).



**Figure 5.** "Present-day" mean (a) air-sea  $\text{CO}_2$  fluxes ( $\text{mmol C m}^{-2} \text{d}^{-1}$ ), (b)  $p\text{CO}_2^{\text{SEA}}$  (ppmv), (c) SST ( $^{\circ}\text{C}$ ), and (d) primary production ( $\text{g C m}^{-2} \text{d}^{-1}$ ) in the Mid-Atlantic Bight (shaded area indicates the standard deviation, and red dots indicate the observations from Takahashi *et al.* [2009]).



**Figure 6.** “Present-day” mean (a) air-sea CO<sub>2</sub> fluxes (mmol C m<sup>-2</sup> d<sup>-1</sup>), (b) pCO<sub>2</sub><sup>SEA</sup> (ppmv), (c) SST (°C), and (d) primary production (g C m<sup>-2</sup> d<sup>-1</sup>) in the South Atlantic Bight (shaded area indicates the standard deviation, and red dots indicate the observations from Jiang et al. [2008]).

using property-property relationships developed by Lee et al. [2000] for DIC and by Millero et al. [1998] for alkalinity, with temperature and salinity data collected between 1981 and 1998 and between 1980 and 1986, respectively. What we believe we are seeing in our results for the GOM is the sensitivity of this region to these fields and how the Lee and Millero relationships are based on databases that do not have sufficiently high sampling in the GOM region to capture its distinct water mass properties. This has highlighted the need for a regionally specific initialization in the Gulf of Maine with respect to DIC and alkalinity, something that is currently being investigated beyond this study.

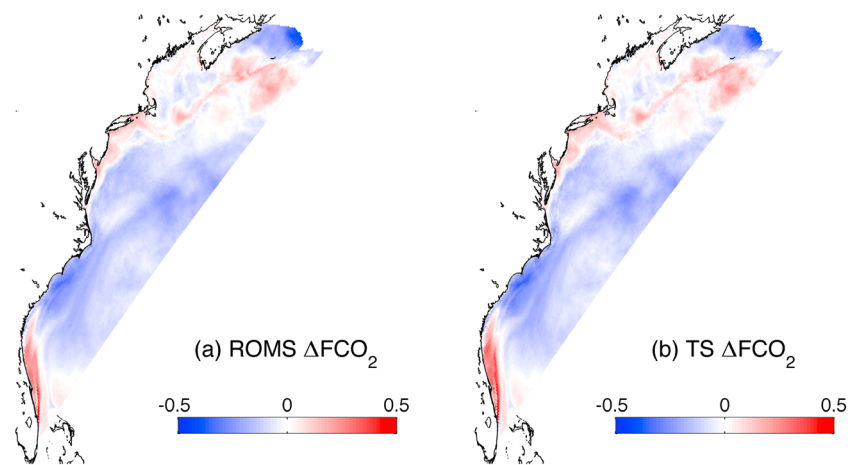
**4.2. Future-Perturbed Air-Sea CO<sub>2</sub> Fluxes and Sensitivity Analysis**

The response of the air-sea CO<sub>2</sub> flux to future-perturbed variations in atmospheric forcing (Figure 3b) is evaluated by examining the magnitude of the difference between the 4 year mean fluxes for each model scenario (Figure 7a). The intention here is to remove the long-term climate variability and focus just on the changes that occur with long-term climate change as represented by the RegCM3-derived atmospheric anomalies. The main differences in the future-perturbed atmospheric forcing fields are found in the air temperature and wind forcing. There is an overall increase in air temperature over the entire model domain ranging from

**Table 2.** Interannual “Present-Day” Air-Sea CO<sub>2</sub> Fluxes (mol C m<sup>-2</sup> yr<sup>-1</sup>) (Positive Flux Indicates That the Ocean Acts as a Source of CO<sub>2</sub> to the Atmosphere, and Negative Flux Indicates That the Ocean Acts as a Sink of CO<sub>2</sub> From the Atmosphere)

Region	Area (10 <sup>11</sup> m <sup>2</sup> )	Air-Sea CO <sub>2</sub> Flux (mol C m <sup>-2</sup> yr <sup>-1</sup> )					Mean (STD)	Obs
		2004	2005	2006	2007			
GOM	11.5	-2.8	-1.3	-1.3	-1.7	-1.8 (0.7)	0.3 <sup>a</sup>	
MAB	8.6	-1.4	-1.0	-1.1	-1.4	-1.2 (0.2)	-1.8 <sup>b</sup>	
SAB	9.2	-0.6	-0.8	-0.4	-0.3	-0.5 (0.2)	-0.5 <sup>c</sup>	

<sup>a</sup>Vandemark et al. [2011].  
<sup>b</sup>Takahashi et al. [2009].  
<sup>c</sup>Jiang et al. [2008].



**Figure 7.** Magnitude of “future-perturbed”-“present-day” (a) ROMS annual air-sea CO<sub>2</sub> flux difference,  $\Delta FCO_2$  (mol C m<sup>-2</sup> yr<sup>-1</sup>), and (b) first-order Taylor series approximation of annual air-sea CO<sub>2</sub> flux difference,  $\Delta FCO_2$  (mol C m<sup>-2</sup> yr<sup>-1</sup>).

1°C offshore to as much as 3°C along coastal regions (Figure 2a). There is also a decrease in the northeasterly wind component in the Gulf of Maine and northern Mid-Atlantic Bight, an increase in the easterly wind component in the southern Mid-Atlantic Bight, and an increase in the southeasterly wind component in the South Atlantic Bight (Figure 2b).

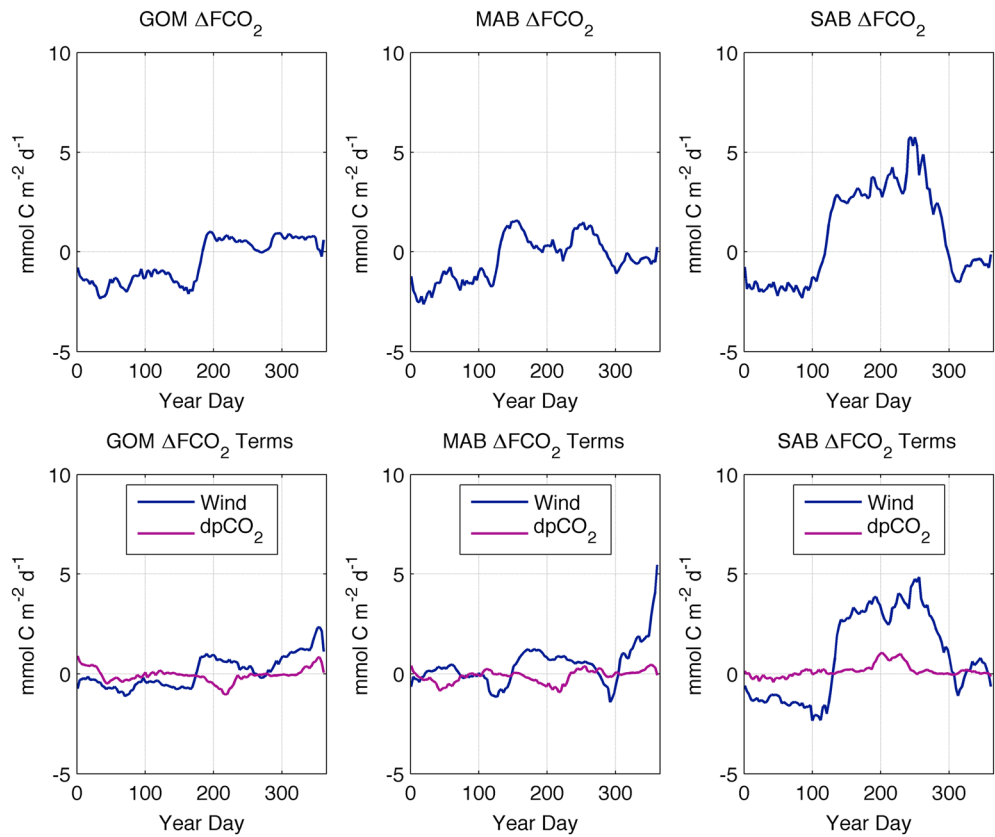
Overall, the most notable spatial differences to the air-sea CO<sub>2</sub> flux in response to the future-perturbed scenario occur in the inner Mid-Atlantic Bight and the southern South Atlantic Bight and to a much lesser extent, the southern coastal Gulf of Maine, where the magnitude of the future-perturbed scenario CO<sub>2</sub> flux is larger than that of the present day (Figure 7a). In contrast, in the inner shelf of the northern South Atlantic Bight, the outer Mid-Atlantic Bight shelf, and to a lesser extent, the northern coastal and outer reaches of Gulf of Maine, the magnitude of the future-perturbed scenario CO<sub>2</sub> flux is less than that of the present day. When the flux differences are integrated over each of the subregions, spatial differences tend to cancel each other out in the Gulf of Maine and Mid-Atlantic Bight but not in the South Atlantic Bight where a significant change occurs. The position of the along-shelf gradient in the sign and magnitude of the air-sea CO<sub>2</sub> flux moves further north, and the subregion as a whole shifts from being a mild sink for atmospheric CO<sub>2</sub> (−0.6 Mt C yr<sup>-1</sup>) to becoming a source of CO<sub>2</sub> (0.7 Mt C yr<sup>-1</sup>) to the atmosphere in response to the future-perturbed scenario (Figure 3b and Table 3).

The Taylor series decomposition analysis highlights the relative contributions of the air-sea CO<sub>2</sub> flux terms to the change in the flux in CO<sub>2</sub>,  $FCO_2$ . Figure 7b shows that the first-order Taylor series estimate of the magnitude of the difference between the future-perturbed and present-day scenarios,  $\Delta FCO_2$ , is a good approximation of the modeled estimates (Figure 7a). The absolute mean difference between Figures 7a and 7b is 6.9 mmol C m<sup>-2</sup> yr<sup>-1</sup>.

Examination of the seasonal contribution of the different wind and  $pCO_2^{SEA}$  flux terms provides some insight into which processes may be important for each of the regions in driving the seasonality of air-sea CO<sub>2</sub> fluxes (Figure 8). Contributions from the Schmidt number and the solubility of CO<sub>2</sub> to  $\delta F$  are small and cancel each other out (not shown).

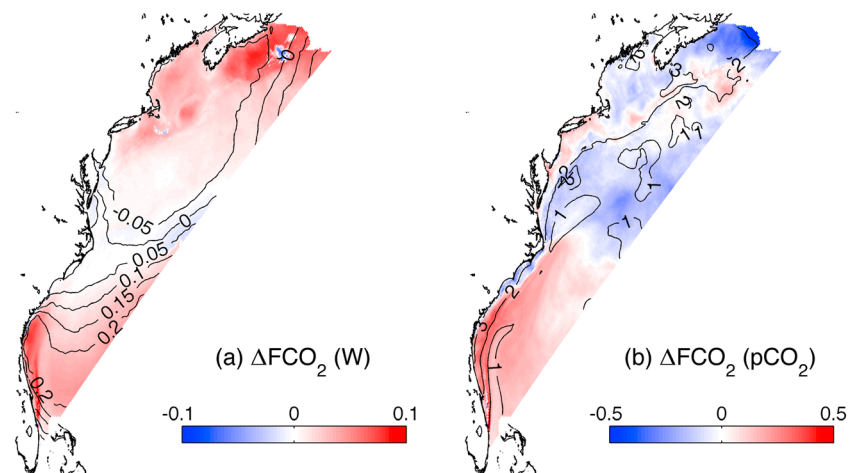
**Table 3.** Four-Year Mean “Present-Day” and “Future-Perturbed” Air-Sea CO<sub>2</sub> Fluxes (mol C m<sup>-2</sup> yr<sup>-1</sup> (Mt C yr<sup>-1</sup>))

Region	Area (10 <sup>11</sup> m <sup>2</sup> )	Air-Sea CO <sub>2</sub> Flux (mol C m <sup>-2</sup> yr <sup>-1</sup> (Mt C yr <sup>-1</sup> ))		
		Present Day	Future Perturbed	Difference
GOM	11.5	−1.8 (−2.5)	−1.7 (−2.4)	0.1 (0.1)
MAB	8.6	−1.2 (−1.0)	−1.2 (−1.2)	0.0 (−0.2)
SAB	9.2	−0.5 (−0.6)	0.2 (0.7)	0.7 (1.3)

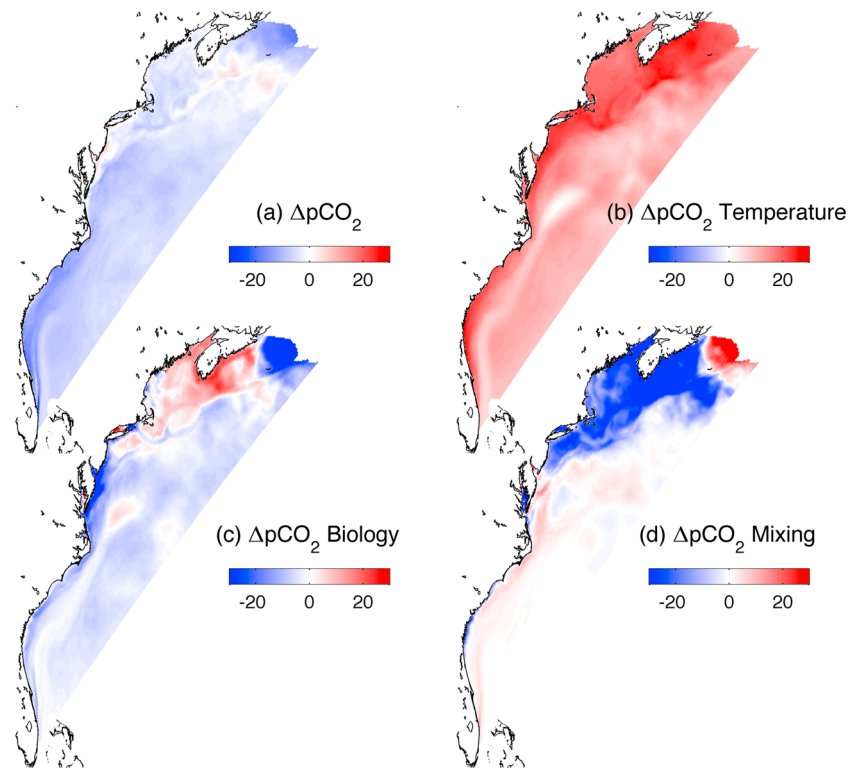


**Figure 8.** (top row) First-order Taylor series approximations of the magnitude of the mean annually integrated regional flux differences. (bottom row) Magnitude of the mean annually integrated regional flux differences due to changes in  $pCO_2^{SEA}$  (red) and flux differences attributable to near-surface (10 m) wind speed changes (blue).

Our analysis of the  $pCO_2^{SEA}$  and wind flux terms shows that it is the  $pCO_2^{SEA}$  that is dominating the annual  $FCO_2$  response to the future-perturbed scenario, with a lesser contribution from winds (Figure 9). However, winds are playing a stronger seasonal control (Figure 8). This is particularly evident in the South Atlantic Bight in spring and summer when higher sea surface temperatures (SSTs) (acting to increase  $pCO_2^{SEA}$ ) combined with higher winds contribute to increased outgassing in the region (Figure 3b). That  $pCO_2^{SEA}$



**Figure 9.** First-order Taylor series approximation of the magnitude of the difference between “future-perturbed” and “present-day” fluxes,  $\Delta FCO_2$  ( $mol\ C\ m^{-2}\ yr^{-1}$ ), (a) magnitude of the annually integrated flux difference attributable to near-surface (10 m) wind speed changes; contours denote the actual annual wind speed difference. (b) Magnitude of the annually integrated flux difference due to changes in  $pCO_2^{SEA}$ ; contours denote the actual SST differences.



**Figure 10.** (a) Second-order Taylor series approximation of  $\Delta p\text{CO}_2^{\text{SEA}}$  ("future-perturbed"-"present-day"), (b) relative contribution of variations in temperature to  $\Delta p\text{CO}_2^{\text{SEA}}$ , (c) relative contribution of variations in biology to  $\Delta p\text{CO}_2^{\text{SEA}}$ , and (d) relative contribution of variations in mixing to  $\Delta p\text{CO}_2^{\text{SEA}}$ .

dominates the annual response over the entire model domain is not unexpected, given the characteristics of the atmospheric anomalies driving the scenario; i.e., the increase in air temperature will drive an increase in SST which acts to increase  $p\text{CO}_2^{\text{SEA}}$ , thus reducing the uptake of  $\text{CO}_2$  by the ocean. Given that our focus here is only on changes in atmospheric forcing and does not take into account a projected future ocean biogeochemical state nor a higher atmospheric  $p\text{CO}_2$ , the adjustment of  $p\text{CO}_2^{\text{SEA}}$  is a direct response to an increase in SST. However, changes in SST will also have an impact on biological activity and mixing of water masses with different DIC/TA characteristics.

In order to understand the changes to  $p\text{CO}_2^{\text{SEA}}$  which are contributing to  $\delta F$ , we further analyze the Taylor series approximations of the difference in  $p\text{CO}_2^{\text{SEA}}$  due to differences in SST, sea surface salinity, biological activity, and the ratio of mixing of water masses with different DIC/TA characteristics to air-sea  $\text{CO}_2$  fluxes (Figure 10). We examine the second-order approximations of  $p\text{CO}_2^{\text{SEA}}$  in this instance because the effects of the different processes that control  $p\text{CO}_2^{\text{SEA}}$  depend on the state of the system.

The negative  $p\text{CO}_2^{\text{SEA}}$  anomalies (Figure 10a) are, for the most part, a clear response to the increase in SST in the future-perturbed perturbation. The contribution of sea surface salinity to  $p\text{CO}_2^{\text{SEA}}$  is not significant (not shown). The relative contribution of biological activity to  $p\text{CO}_2^{\text{SEA}}$  (Figure 10c) is an indication of the effects of anomalous biological activities on DIC and TA pools. It can be interpreted as a sign of net ecosystem production (NEP; net primary production—respiration), in other words, the rate of organic carbon accumulation. Areas of enhanced NEP are associated with a negative biological contribution to the annual mean  $p\text{CO}_2^{\text{SEA}}$  difference, while areas of reduced NEP are associated with a positive biological contribution to the annual mean  $p\text{CO}_2^{\text{SEA}}$  difference [Previdi *et al.*, 2009]. The relative contribution of the ratio of mixing to air-sea  $\text{CO}_2$  fluxes on the annual mean  $p\text{CO}_2^{\text{SEA}}$  difference (Figure 10d) effectively balances that from biological activity (Figure 10c). Increased biological production will have the effect of drawing down surface water DIC and decreasing surface  $p\text{CO}_2^{\text{SEA}}$ , thus enhancing atmospheric  $\text{CO}_2$  uptake which will counteract the effects of enhanced production. It is the combination of all these three processes (i.e., net effect of biological activity, the ratio of mixing to air-sea  $\text{CO}_2$  fluxes, and variability in SST) that is driving the variation in  $p\text{CO}_2^{\text{SEA}}$ . Overall,

we find an approximately 1:1 ratio between temperature and the net effect of biology and mixing, indicating the importance of both processes in controlling the  $p\text{CO}_2^{\text{SEA}}$  variability in our future-perturbed scenario.

## 5. Discussion and Conclusions

Our assessment of air-sea  $\text{CO}_2$  fluxes for three subregions along the U.S. East Coast continental shelf region indicates that annually, all subregions act as a sink for atmospheric  $\text{CO}_2$ , with some interannual variabilities. The Gulf of Maine exhibits the largest interannual variability, possibly in response to NAO-driven variability in atmospheric forcing.

There is a clear along-shelf gradient in air-sea  $\text{CO}_2$  fluxes (south to north) along the whole shelf region with estimates ranging from  $-0.5 \text{ Mt C yr}^{-1}$  in the South Atlantic Bight to  $-1.0 \text{ Mt C yr}^{-1}$  in the Mid-Atlantic Bight and  $-2.5 \text{ Mt C yr}^{-1}$  in the Gulf of Maine. Seasonality in the fluxes is evident, with autumn and winter being the strongest  $\text{CO}_2$  sink periods, corresponding with lower  $p\text{CO}_2^{\text{SEA}}$  values driven by lower surface temperatures. During the summer months (June to August), some outgassing occur when surface warming tends to increase  $p\text{CO}_2^{\text{SEA}}$ , thus decreasing the uptake of atmospheric  $\text{CO}_2$ .

The sensitivity of air-sea  $\text{CO}_2$  fluxes to variability in atmospheric forcing shows that an increase in air temperature of up to  $3^\circ\text{C}$  in the atmospheric anomalies can have important spatial consequences for the relative contribution of  $p\text{CO}_2^{\text{SEA}}$  to the air-sea  $\text{CO}_2$  flux response. To a lesser extent, the increase in northeasterly and southeasterly wind components in the future-perturbed perturbation also had important spatial consequences for the air-sea  $\text{CO}_2$  flux response in the Gulf of Maine and South Atlantic Bight, respectively.

Spatial differences tend to cancel each other out when the fluxes are integrated over each of the subregions, except in the South Atlantic Bight where a significant change occurs in response to the future-perturbed perturbation. The position of the along-shelf gradient moves further north, and the subregion experiences a regime shift from being a mild sink for atmospheric  $\text{CO}_2$  ( $-0.6 \text{ Mt C yr}^{-1}$ ) to becoming a source of  $\text{CO}_2$  ( $0.7 \text{ Mt C yr}^{-1}$ ) to the atmosphere.

The net effect of variability in biology and the mixing/air-sea  $\text{CO}_2$  flux ratio combined with variability in SST are the important processes driving variability in  $p\text{CO}_2^{\text{SEA}}$ . This is consistent with *Signorini et al.* [2013] who showed that changes in DIC and SST are the main factors driving seasonality in  $p\text{CO}_2^{\text{SEA}}$ . *Vandemark et al.* [2011] found that the net effect of biological activity and the ratio of mixing to air-sea  $\text{CO}_2$  fluxes was the greater control over variations in  $p\text{CO}_2^{\text{SEA}}$  in late March and into the summer in the Gulf of Maine. Later in the year, temperature and the net effect of biology and mixing were equivalent, indicating that throughout the year, solubility is in competition with other biological and mixing  $p\text{CO}_2^{\text{SEA}}$  controls in the Gulf of Maine. *Jiang et al.* [2008] found a 1:1 ratio between water temperature and all other  $p\text{CO}_2^{\text{SEA}}$  dynamics throughout the year in the South Atlantic Bight and concluded that temperature is the dominant control of  $p\text{CO}_2^{\text{SEA}}$  variability in the South Atlantic Bight. *DeGrandpre et al.* [2002] found a similar temperature-dominated control over  $p\text{CO}_2^{\text{SEA}}$  variability in the Mid-Atlantic Bight. We found that temperature was the dominant control of future-perturbed  $p\text{CO}_2^{\text{SEA}}$  variability for all subregions.

It is likely that our NENA model scenarios are not capturing the complex springtime and late autumn dynamics that regulate  $p\text{CO}_2^{\text{SEA}}$  in the Gulf of Maine. Thus, they are not resolving the competition between solubility and biology/mixing controls in the  $p\text{CO}_2^{\text{SEA}}$  variability in this region. We suppose that the model is not capturing important changes in water mass properties during springtime and winter transition periods because our initialization of DIC and alkalinity in the Gulf of Maine does not include sufficient samples from the Gulf of Maine, highlighting the need for a regionally specific initialization in the Gulf of Maine with respect to DIC and alkalinity. More generally, changes to the water mass properties over the last 40 years, especially a warming and freshening trend in this subregion as a result of climate change, have been reported as virtually certain by the latest IPCC assessment [*Rhein et al.*, 2013], suggesting the need to revisit the fitness for purpose of the *Lee et al.* [2000] and *Millero et al.* [1998] regional relationships.

It is worth remembering that our study focuses only on the sensitivity of shelf air-sea  $\text{CO}_2$  fluxes to perturbations in atmospheric forcing; however, understanding what drives the response of air-sea  $\text{CO}_2$  fluxes in shelf waters, in particular in the context of climate change, is in reality more complicated than this. Our model does not resolve the detailed structure of inland and marsh waters nor does it resolve the complex biogeochemical

transformations that take place in rivers and estuaries prior to reaching the coastal ocean. Nevertheless, we recognize that the impact of changes to riverine inputs is an extremely important and complex problem in itself and the subject of several recent studies. Tao et al. (personal communication) show that river discharge in the region is projected to increase by 40–53% (16–30%) for the IPCC A2 (B1) scenarios by the end of this century. Herrmann et al. [2015] show that 60% of the organic carbon entering the U.S. East Coast estuaries actually makes it out to the shelf, while Laruelle et al. [2015] highlight how spatial variability in the efficiency of carbon removal through the estuarine filter contributes to the magnitude of the spatial gradient of CO<sub>2</sub> sinks along continental shelf waters of the U.S. East Coast. Their study also reveals that ice and snow cover are important controlling factors of the seasonal dynamics of CO<sub>2</sub> outgassing in rivers and account for significant spatial differences between the Gulf of Maine and Mid-Atlantic Bight regions.

Feng et al. [2015] show that only a fraction of the inorganic nitrogen that enters rivers actually makes it to the shelf and that this fraction has changed a lot over time due to climate change and land use change. Moreover, many of the riverine changes are due to land use changes and it is very difficult to separate the effects of climate change and land use change. Tian et al. [2015] have looked at historical changes (1901 to 2008) in riverine carbon inputs specifically for the GOM, MAB, and SAB regions and find significant changes in the DIC flux. In the GOM, they find an increasing DIC trend, while in the MAB and SAB, they find a decreasing DIC trend. No significant change was found in the organic carbon riverine inputs in all regions. Clearly, the impact of climate change on riverine inputs, especially changes to the freshwater input and transport of carbon into the coastal ocean, adds an important layer of complexity to understanding the air-sea flux of CO<sub>2</sub> on the U.S. East Coast continental shelf.

On decadal time scales, studies of the global coupled carbon-climate system suggest that the ocean may become less efficient sink for anthropogenic CO<sub>2</sub> due to positive feedback in the coupled carbon-climate system [Gruber et al., 2004]. The amount of DIC in seawater for a given atmospheric partial pressure and ocean total alkalinity is highly temperature dependent. Gruber et al. [2004] estimate that a surface ocean warming of 2°C and a 0.5°C warming over the upper 300 m in the next 20 years could lead to an equilibrium loss of about 15 Pg C. Moreover, increases in stratification as a result of increases in upper ocean temperature equivalent to 1.5°C can lead to a reduction in the cumulative uptake of CO<sub>2</sub> by the ocean over 20 years on the order of 10%.

Our study provides a view on how the local response of air-sea CO<sub>2</sub> fluxes to a surface ocean warming of between 2°C and 3°C can differ significantly between subregions. It underlines the role of temperature in regulating  $p\text{CO}_2^{\text{SEA}}$  variability and the complex feedback between solubility and biology/mixing controls. Whereas the South Atlantic Bight shows a shift from being a sink of CO<sub>2</sub> to being a source of CO<sub>2</sub> in its annual air-sea CO<sub>2</sub> flux as a result of surface warming, the Gulf of Maine and Mid-Atlantic Bight show no difference in their annual air-sea CO<sub>2</sub> fluxes. In the South Atlantic Bight where the present-day scenario estimates it as being weak sink of CO<sub>2</sub>, variability in winds plays a stronger seasonal control over  $p\text{CO}_2^{\text{SEA}}$  in spring and summer when higher SSTs (acting to increase  $p\text{CO}_2^{\text{SEA}}$ ) combined with higher winds are most likely contributing to net annual outgassing in the future-perturbed scenario. While the time frame of our study is too short to qualify as a climate study, it does provide a preliminary indication of the scale of changes to air-sea CO<sub>2</sub> fluxes that may occur as a result of global warming along the U.S. East Coast continental shelf subregions.

#### Acknowledgments

We would like to thank Dave Pollard for supplying the RegCM3 model output which we used to perturb our future-perturbed scenario and Mike Previdi for providing essential guidance and MATLAB scripts with respect to the Taylor Series decomposition analyses. B.C. would also like to thank Charlie Stock for some very helpful discussions of an early draft of the paper. We gratefully acknowledge the sponsors of the project, the NASA Ocean Biology and Biogeochemistry Program, with funding from both the Interdisciplinary Science and Carbon Cycle Science Program. Finally, we would like to thank two anonymous reviewers whose constructive comments helped to significantly improve our manuscript. Output of the ROMS scenarios used in our analysis is available with unrestricted access from a Rutgers University via Thematic Real-Time Environmental Distributed Data Services (THREDDS) at <http://tds.marine.rutgers.edu/thredds/roms/catalog.html>. Contact John Wilkin ([wilkin@marine.rutgers.edu](mailto:wilkin@marine.rutgers.edu)) for the guidance on which THREDDS entries correspond to the scenarios presented here.

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