Author Response for Major Revisions to bg-2015-408

Climate impacts on multidecadal pCO_2 variability in the North Atlantic: 1948-2009

Melissa L. Breeden* and Galen A. McKinley

Department of Atmospheric and Oceanic Sciences, University of Wisconsin, Madison, Wisconsin, USA

***Corresponding author: mbreeden@wisc.edu; 1225 W. Dayton St. Madison, WI 53706**

Editor Decision and Author Response

Associate Editor Decision: Reconsider after major revisions (12 Jan 2016) by Prof Dr Leticia Cotrim da Cunha
Comments to the Author:
Dear Dr. Breeden,

Thank you for fully addressing all reviewers' comments.
I strongly suggest you to submit a revised manuscript version where all changes made to your work are highlighted (italic, bold, coloured font), in accordance with your reply.
For instance, according to your reply to Q1, Reviewer 2: please show that you have expanded section 2.1 to improve model description in your manuscript, and so on. This will certainly help the reviewers.

I have just one quick question: in your reply to Reviewer 1, you mentioned a revision to Figure 2 (original manuscript), but in the end of your document it appears as fig. "1". I have compared with the manuscript's first version, and it is ok, but maybe you could mention this to the reviewer, ok?

Thank you again, and I am looking forward to reading your revised manuscript.
Leticia Cotrim da Cunha

Non-public comments to the Author:
Dear Dr. Breeden and Dr. McKinley

Thank you again for submitting your manuscript to BGD. I would like to apologise for the delay, but I was still expecting to receive, together with your rebuttal, a revised manuscript version. Somehow I didn't completely understand the new rules for publishing, I am sorry.
I have decided to reconsider after major revisions so that the referees will be able to read a revised version of your manuscript, and we'll then proceed to a final decision.
With my best wishes
Leticia Cotrim da Cunha

Dear Dr. Cotrim da Cunha,

We are pleased to present the requested revised manuscript and revised supplementary material as per your request. We understand there have been recent changes to the procedure for publishing, which now explicitly states not to prepare the revised manuscript as part of responding to reviewer comments. We are happy to provide the revised documents for the ease of you and the reviewers.

This document contains the following:

1. Copies of the responses to reviewers 1 and 2 that were submitted in December
2. A list of key changes and reorganization of the documents
3. Revised manuscript with changes marked
4. Revised supplementary material with changed marked

We hope this will provide the information needed to reach a decision concerning our manuscript. We appreciate the time you have taken to help the revision of this paper.

Sincerely,
Melissa Breeden
Galen McKinley

1. Response to Reviewers

Reviewer #1 Major Comments

1) SST and DIC have been identified as the variables of interest. This seems sensible, but because much of the analysis is qualitative (in the sense that patterns are compared with each other, and unit-less time series are compared with each other), the fact that these patterns and timeseries typically match with the explanatory variables does not allow us to definitively accept DIC and SST as the drivers. The analysis (e.g. figure 1 and 2) should also be carried out with the other important candidate driver, alkalinity. Alkalinity may well be important, and for the narrative in the paper to hold up, this needs to be either ruled out, or brought into the story.

1. Thank you for this helpful suggestion. We have included EOF analysis of the alkalinity contribution to pCO_2 in Figure 2 (below). EOF1 explains 19% of the variance of the pCO_2-ALK, and has a center of maximum change in the subpolar gyre. The principal component (PC1) of this pattern, however, does not correlate highly to PC1's of total pCO_2, and pCO_2-chem (r=-.25; r=.44, respectively), or to the AMO (correlations added to the supplementary table 1). This indicates that while alkalinity does have a contribution to the spatial pattern shown in the EOF1 of pCO_2-chem, the temporal evolution differs substantially. Conversely, the primary mode of DIC variability, captured in the EOF1 of pCO_2-DIC, does correlate strongly with AMO and with pCO_2-chem itself. Discussion to this effect will be included in the revised manuscript.

Further, we hope to bring to your attention that the analysis here is not, as suggested, simply qualitative. The variability of pCO_2 and its components is quantified in units of uatm (Fig 1-3) and for DIC units of mmol/m3 (Fig 4-5). Variability over time is quantified as the value of the map multiplied by the unitless timeseries.

2… In this simulation, the authors use a realistic atmospheric forcing, therefore if they were to widen their definition of the AMO to include the idea (that is gathering weight) that a substantial component of the AMO variability over the interval of interest could be atmospherically forced (rather than resulting from internal ocean variability) - see Booth et al., Nature 2012 - it might be possible to justify the narrative presented here even if the AMOC changes don't fit with those many would suggest are intimately associated with the AMOC variability.

2. Yes, we agree that consideration of the MOC in the model is important for this analysis. We show the MOC time series in the supplementary material (Figure S6) and find that it is not directly correlated to the AMO, but rather covaries with the NAO (supplementary Table 1,2). This is consistent with Booth et al. (2012) who suggest that the AMO may be driven more by atmospheric aerosol forcing than internal oceanic variability as represented by the MOC. We will include this discussion and citation in the revised text.

Further, we will emphasize in the revised text that since this model is a regional model that is restored to climatological T and S at 20S (as clarified in the text in response to Reviewer 2 comments), it should be primarily atmospheric forcing that generates the model AMO. Mechanisms of internal ocean variability involving the Southern Ocean are not present here.

3… I would like to have confirmed that the regressions onto the AMO definitely relate to the AMO 'down-and-up', rather than (e.g.) the AMO's trend. Because the AMO is higher in 2009 than 1948, if any of the factors that are look at in figures 4 and 5 also have some trend, the regression could pick this up even if the multidecadal variability were not playing a role. A simpler to understand and more robust figure (in my opinion) would just present difference maps between the high-AMO periods and low-AMO period with everything first detrended. If the vertical mixing narrative presented in the paper definitely does explain the time series in figure 2, this should be very clear in these plots.

3. What the reviewer describes as his/her desire is precisely what the regression plots in Figures 4 and 5 provide. These fields illustrate the change in each field at each grid point associated with an AMO index of one standard deviation. To consider the change in any field when the AMO has a value of 2 as observed at the end period, one should double the regression pattern, and so forth. This method does not allow the higher value of the AMO at the end of the period to dominate over other periods of time. Additional confirmation can be found with Supplementary Figure 2 in which typically low AMO period (1970-80s) are compared to the typically high AMO periods (1950-60s and 1990-2000s). These illustrate patterns that are quite comparable to the regressions.

4) Finally, it is pretty important to rule out any contributions here from model drift. The authors state that 'drift in the biogeochemical parameters is eliminated' after a 60 year spin-up. Perhaps I'm overly skeptical, but find this somewhat hard to believe - 60 years is a very short spin-up. Can the authors present evidence for this, or present data from a parallel control run (if this exists)? As noted above, drift could really influence this analysis.

4. Thank you for this concern, as it has caused us to return to our notes to confirm the length of the biogeochemical spinup. The physical model is spun up for 100 years before the biogeochemical parameters are introduced and spun up with biogeochemistry for, in fact, 100 years. The percent change over the last five years of spinup in the basin-averaged surface DIC field is 0.00046% per year. For comparison, the percent change in DIC from a high AMO (1955) to low AMO (1975) is .012% per year, two orders of magnitude greater than drift at the end of the spin up. Therefore we do believe that a 100-year biogeochemical spinup is sufficient to eliminate model drift in the upper ocean, which is the region of focus for this study. We will include this updated information and change comparison in the revised text.

Reviewer #1 Minor Comments

1) I'm not convinced that figure 3 is particularly useful. Is column 1 not essentially just column 3 minus column 2 (based on the definition of the AMO)? In which case, I found the explanation built around this figure overly complicated. I wonder if this could be removed, and the points made with reference to this figure be made instead by just contrasting figures 1 and 2?

Minor 1. We appreciate your concern about potential redundancy in the figures and do recognize that the resulting structures have many similarities. The important distinction here is how the patterns are derived. EOF analysis objectively identifies the spatial pattern explaining the greatest fraction of variability in a field, and also returns how this pattern varies in time with the principle component associated with each EOF spatial field. Though in several cases these principle components have strong correlations with the AMO or SST trend, they are not strictly identical to these indices. It is, however, of strong interest that the dominant mode of variation in pCO_2-DIC, in particular, is the AMO without this index being given to the statistical analysis.

Regression analysis, in contrast to the EOF approach, prescribes an index of temporal variation and retrieves the associated response to this index in each pCO_2 field. With this technique we are able to isolate the pCO_2 and component responses to the AMO (Figure 3, column 1) as separate from the SST trend and to the total SST pattern (Column 2,3) . This distinction is not possible using EOF analysis alone. It is a point we hope to make that these patterns combine in space, and will clarify the text. But we do not feel that these points can be clearly made without Figure 3.

2) I like the use of the barotropic stream function and MLD changes to explain the vertical DIC changes. I wonder if it might be useful to move these into the main paper? I would also suggest it is worth pointing out that the changes are broadly in

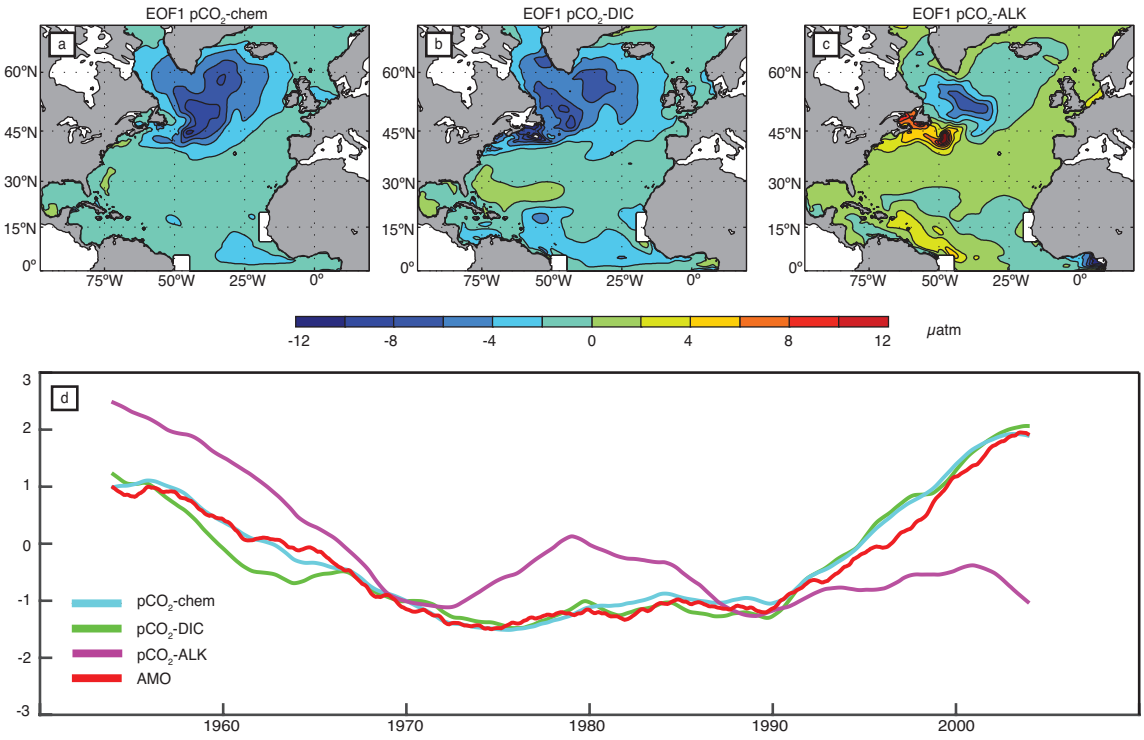
agreement with the observed changes (e.g. Zhang, 2008, GRL).
Minor 2. Thank you, we agree that these figures are important to the text and shall include them in the main text. We shall add the suggested reference as well.

3) It would be useful if the methodology section could include an explanation of why a regional model was used, and some basic model validation. Currently the only validation that I can see relates to the temporally and spatially averaged N. Atlantic CO2 uptake. Perhaps this is published elsewhere?
Minor 3. Thank you, we have added model validation in response to the comments from Reviewer 2 – please see below. We chose a regional model because we are focused on North Atlantic carbon variability specifically and to reduce the computational requirements for this goal the by limiting the model's geographical scope. Also, as referenced, previous work (Ullman et al., 2009, Bennington et al. 2009) has shown that this model performs well enough to interrogate processes related to North Atlantic carbon variability

5) NASST not defined as far as I can see
Minor 5. NASST is defined in Section 2.2 as the total North Atlantic SST index.

6) There are a few minor wording issues that will hopefully be picked up in a revised manuscript - e.g. 'of' missing P15225 line 14-15, trend(s) P15224 line 9. . .
Minor 6. Thank you for noting these typos, we will eliminate these errors in the revised manuscript.

Revised Figure 2



Top left: EOF1 pCO_2-chem (µatm), top middle: EOF1 pCO_2-DIC (µatm), top right: EOF1-pCO2-ALK, explaining 32%, 25% and 19% of total variance, respectively, Bottom: PC1- pCO_2-chem (dark blue), PC1- pCO_2-DIC (green) and AMO index (red), all standardized. Timeseries smoothed with a 121-month box smoother.

Reviewer #2

I find the writing of the manuscript misses much of the necessary details to assure reproducibility. I suggest the authors to consider significantly expanding the section 2.1 and/or provide technical details in the appendix in order to adequately document what went into this calculation, and if any, please discuss significant changes in model parameters and/or bcs from the earlier work.

Thank you for pointing this out. As requested, we have expanded section 2.1 with additional details on the model, and also emphasize the reference to Ullman et al. 2009 and Bennington et al. 2009 that describe this model. We will proceed to include the following details, as requested:

What is the timescale used for the SST and SSS relaxation?

SST with a timescale of 2 weeks; SSS with a timescale of 4 weeks.

Are you using the glacier melt and/or river discharge to force the model salinity?

Glacier melt and/or river discharge are not included in the model forcing, instead the SSS relaxation approximates these impacts.

Are the freshwater forcing consistent between salinity and tracers?

Yes, the E-P forcing and SSS relaxation impacts both salinity and tracer concentrations.

How is the sea-ice dynamics treated?

Fractional ice from NCEP Reanalysis 1 is applied, with interpolation to daily.

How are the open boundary conditions set?

A sponge layer exists at 20S, and over the first 5 degrees of latitude to the North, there is restoration to climatological T, S, DIC and phosphate fields. For T and S, there is also a sponge layer at Gibraltar. More discussion of the sponge layer can be found in Ullman et al. 2009.

I think this is an important problem for calculating pCO2, but maybe the authors can explain what's important for CO2 and how well the model captures it in the N Atlantic.

This simulation is essentially identical to that of Ullman et al. 2009 and Bennington et al. 2009, except for the longer simulation period and the pre-industrial pCO2 that is applied. Given that the surface ocean data to which we can compare has the influence of anthropogenic pCO2, we rely on the extensive biogeochemical comparisons to BATS and subpolar gyre DIC and ALK and pCO2 and SST variability

and trends. Here, we compare to the natural carbon uptake estimate from Mikaloff-Fletcher et al. 2007, and include the subpolar gyre DIC profile compared to GLODAP (see Figure at bottom) in the supplementary.

There are many zonal and meridional WOCE/Clivar transects in the time period following 1990s. As the analysis of DIC variability (3.3) emphasizes the importance of the vertical mixing, it would be good to show how well this model reproduce the vertical gradients of DIC and alkalinity in the subpolar regions.

As this is a pre-industrial model run, it is not possible to directly compare the model to data collected in the modern era, as of course the ocean DIC concentration has significantly increased over time. Thus, we compare here a 0-2000m 1948-2009 model DIC profile averaged over the subpolar gyre (35-55N, 5-60E) to the GLODAP (Key et al. 2004) estimate of pre-industrial DIC (see Figure at bottom). The model is largely within the uncertainty of the observed estimate from 0-2000m, and is a few percent below the observed estimate from 2000-4000m . As the average annual maximum MLD of this region is 284m, and the annual maximum MLD for all points in the region is, on average, 3215m, this comparison indicates that the model does capture the vertical distribution of pre-industrial DIC in a manner that is consistent with the available observations. This figure will be included in the Supplementary Material.

Line 23 in page 15226. Is the 100+60 year spin up enough? Is there a residual drift in the model at the end of the biogeochemical spin up? If any, please quantify the drift with respect to the variability/trend from the simulation period. Fundamental issue here is that the timescale of AMO is comparable to the simulation lengths itself and also the spin-up length. This would raise reasonable doubt unless clearly justified.

Thank you, this point was also raised by Reviewer 1. Our response to their point 4, quoted here is "4. Thank you for this concern, as it has caused us to return to our notes to confirm the length of the biogeochemical spinup. The physical model is spun up for 100 years before the biogeochemical parameters are introduced and spun up with biogeochemistry for, in fact, 100 years. The percent change over the last five years of spinup in the basin-averaged surface DIC field is 0.00046% per year. For comparison, the percent change in DIC from a high AMO (1955) to low AMO (1975) is .012% per year, two orders of magnitude greater than drift at the end of the spin up. Therefore we do believe that a 100-year biogeochemical spinup is sufficient to eliminate model drift in the upper ocean, which is the region of focus for this study. We will include this updated information and change comparison in the revised text."

Definition of "intense"-ness is vague in the 1st sentence of the abstract. Were you considering per unit area uptake rates? In terms of the integrated carbon uptake, it might be smaller than the SH extra-tropics whose carbon uptake is close to 1PgC/yr.

Thank you, we will specify here that "intense" is in terms of the per-unit-area rate of uptake.

Line 3 in page 15228. Again, please specify whether the freshwater forcing include the E-P from NCEP reanalysis + SSS restoring term.

Thank you, we will include this. E-P from NCEP was included along with the SSS restoring.

Line 25 page 15232, it reads as if the logic is inverted where chemistry controls physics. "The AMO is strongly associated with chemical change" should read like "The AMO strongly influences the chemical change".

We will modify this text.

References in this response:

Key, R. M. *et al.* A global ocean carbon climatology: Results from Global Data Analysis Project (GLODAP). *Global Biogeochem Cy* **18,** GB4031 (2004).

New Figure S1

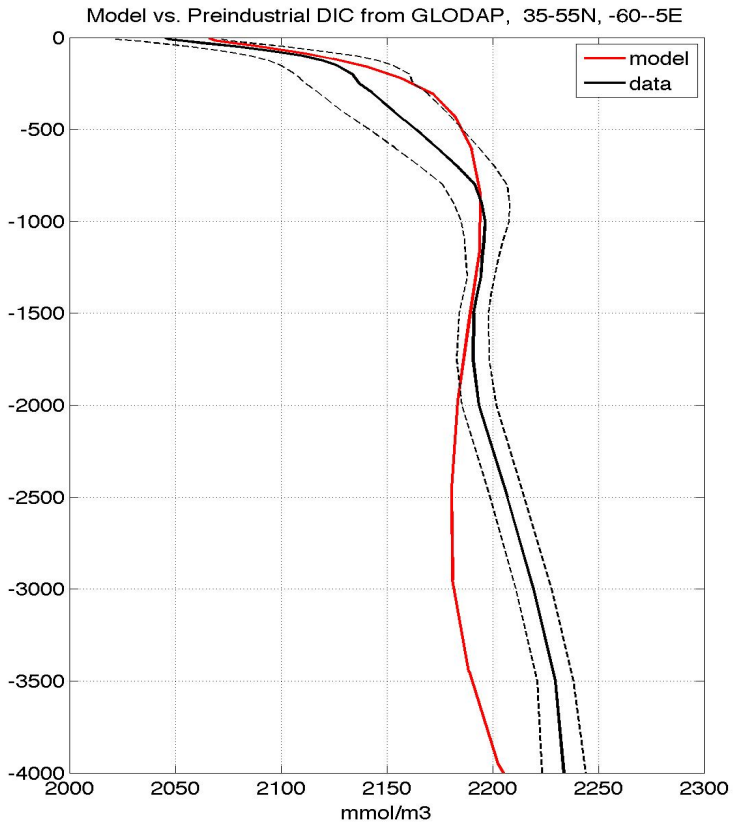


Figure S1: 35-55N, 5-60E average profile of 1948-2009 Model (red) and GLODAP preindustrial DIC (black) in mmol/m^3

2. Key Changes to Manuscript and Supplementary Material

1. Expansion of model details and spinup as requested by both reviewers in section 2.1

2. Addition of model comparison of vertical profile of DIC to GLODAP preindustrial estimates as Figure S1 and complementary supplementary text

3. Modified Figure 2 to include EOF1 of pCO_2-ALK and addition of discussion of alkalinity in section 3.1

4. Figures S2 and S3 moved to main text as Figure 5 and Figure 6 as requested by reviewer #1

5. Included discussion and citations: Zhang (2008) in section 3.3 and Booth et al. (2012) in section 4

6. Modified Table S1 includes correlations with PC1-pCO_2-ALK

Climate impacts on multidecadal pCO_2 variability in the North Atlantic: 1948-2009

Melissa L. Breeden* and Galen A. McKinley

Department of Atmospheric and Oceanic Sciences, University of Wisconsin, Madison, Wisconsin, USA

***Corresponding author: mbreeden@wisc.edu; 1225 W. Dayton St. Madison, WI 53706**

Abstract

The North Atlantic is the most intense region of ocean CO_2 uptake in term of units per area. Here, we investigate multidecadal timescale variability of the partial pressure CO_2 (pCO_2) that is due to the natural carbon cycle using a regional model forced with realistic climate and pre-industrial atmospheric pCO_2 for 1948-2009. Large-scale patterns of natural pCO_2 variability are primarily associated with basin-averaged sea surface temperature (SST) that, in turn, is composed of two parts: the Atlantic Multidecadal Oscillation (AMO) and a long-term positive SST trend. The North Atlantic Oscillation (NAO) drives a secondary mode of variability. For the primary mode, positive AMO and the SST trend modify pCO_2 with different mechanisms and spatial patterns. Positive AMO is also associated with a significant reduction in dissolved inorganic carbon (DIC) in the subpolar gyre, due primarily to reduced vertical mixing; the net impact of positive AMO is to reduce pCO_2 in the subpolar gyre. Through direct impacts on SST, the net impacts of positive AMO is to increase pCO_2 in the subtropical gyre. From 1980 to present, long-term SST warming has amplified AMO impacts on pCO_2.

1 Introduction

To date, the ocean has removed approximately 1/3 of all anthropogenic carbon emitted to the atmosphere and has, thus, substantially damped climate warming (Khatiwala et al., 2009; Sabine et al., 2004). As carbon dioxide emissions continue to increase due to fossil fuel emissions and cement production, there is significant interest in better understanding the ocean carbon cycle. Due to the limited instrumental record and sparse data, multidecadal variability of the ocean carbon sink remains poorly constrained. The North Atlantic, in particular, is a region of highly concentrated carbon uptake (Takahashi et al., 2009) and of significant carbon cycle variability related to variations in the climate, with multiple studies finding an association with the North Atlantic Oscillation (Fay and McKinley, 2013; Schuster et al., 2013; Terry, 2012; McKinley et al., 2011; Loptien and Eden, 2010; Ullman et al., 2009; Thomas et al., 2008). However, data are sparse, processes are complex and the timescales for studies have differed, and this has complicated a clear elucidation of the mechanisms of North Atlantic carbon cycle variations.

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Schuster et al. (2009) analyzed in situ pCO_2 measurements, and suggested a substantial decline in North Atlantic carbon uptake from the mid-1990's to the mid-2000's. LeQuéré et al. (2010) also interpreted observations and models to conclude that there had been a decline in the North Atlantic sink from 1981-2007 due to changing wind patterns and increasing SST. Metzl et al. (2010) focused on subpolar surface ocean carbon cycle changes between 1993-2008, and also concluded that there had been a reduction in carbon uptake. In situ pCO_2 measurements have also been synthesized to illustrate the strong sensitivities of such changes to the locations and timeframe for the analyses (Fay and McKinley 2013; McKinley et al., 2011). The substantial spatial heterogeneity and temporal variability in the North Atlantic complicates efforts to use sparse observations to quantify carbon uptake. Thus, the magnitude and mechanisms North Atlantic carbon cycle variability remains loosely constrained. The present study takes advantage of the full spatial and temporal coverage of a regional numerical model to gain new insights into the mechanisms of variability of North Atlantic pCO_2.

As shown by Ullman et al. (2009) in a 15-year simulation (1992-2006), internal variability in the North Atlantic is partially obscured by the large, quasilinear trend of CO_2 flux into the ocean that is driven by increasing CO_2 emissions. To examine the carbon sink variability that is partially masked by this large carbon influx, we use a hindcast model from 1948-2009

forced with the preindustrial atmospheric CO_2 concentration and realistic climate. As described below, we find that the basin-average SST is associated with the leading mode of surface ocean pCO_2 variability. This SST signal, in turn, includes an upward trend due to greenhouse gas emissions and a signal of internal variability characterized by the Atlantic Multidecadal Oscillation (AMO, Kerr, 2000).

2 Methodology

2.1 Physical-Biogeochemical-Ecosystem Model

The MIT Ocean General Circulation Model (Marshall et al., 1997a, 1997b) has been regionally configured for the North Atlantic between 20°S and 81.5°N (Bennington et al., 2009; Ullman et al., 2009). The model has a horizontal resolution of 0.5° latitude and 0.5° longitude and 23 vertical levels beginning with a resolution of 10m thickness at the surface and increasing to 500 m thickness at 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 model sub-grid-scale processes. Daily fields from NCEP/NCAR Reanalysis I force the model from 1948-2009 (Kalnay et al., 1996). SST and SSS are relaxed to monthly historical SST (Had1SSTv1.0, Rayner et al., 2003) and climatological SSS (Antonov et al., 2006) observations, on the timescale of two and four weeks, respectively. Glacier melt and/or river discharge are not included in the model forcing, instead the SSS relaxation approximates these impacts. Freshwater (evaporation – precipitation) forcing and SSS relaxation impacts both salinity and tracer concentrations. In lieu of an active sea ice simulation, observed fractional ice from NCEP Reanalysis 1 is applied with interpolation to daily resolution.

For open boundary conditions, a sponge layer exists at 20S, and over the first 5 degrees of latitude to the North, there is restoration to climatological temperature, salinity, DIC and phosphate fields. For temperature and salinity, there is also a sponge layer at Gibraltar. More discussion of the sponge layer can be found in Ullman et al., 2009.

The pelagic ecosystem is parameterized using one zooplankton class and two phytoplankton classes (diatoms and 'small' phytoplankton) as described previously (Dutkiewicz et al., 2005; Bennington et al., 2009; Ullman et al., 2009). Carbon (inorganic and dissolved and particulate organic), alkalinity (ALK), phosphorus, silica and iron cycling are explicitly included in the biogeochemical model. Carbonate chemistry is modeled as in Follows et al. (2006). The objective of this simulation is to identify climate impacts on the natural carbon cycle without the

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Moved up [1]: For open boundary conditions, a sponge layer exists at 20S, and over the first 5 degrees of latitude to the North, there is restoration to climatological temperature, salinity, DIC and phosphate fields. For temperature and salinity, there is also a sponge layer at Gibraltar. More discussion of the sponge layer can be found in Ullman et al., 2009.

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complication of the large CO_2 flux into the ocean that is observed. Thus, atmospheric pCO_2 is fixed at a constant, preindustrial level of 278 ppmv.

The physical model was spun up for 100 years. Following the physical spinup, the biogeochemical model was initialized using preindustrial estimates for DIC and ALK climatology from the GLODAP database (Key et al., 2004). The biogeochemical model was then spun up for an additional 100 years, long enough to eliminate drift in the biogeochemical parameters. The percent change over the last five years of spinup in the basin-averaged surface DIC field is 0.00046% per year. For comparison, the percent change in DIC from a high AMO (1955) to low AMO (1975) is .012% per year, two orders of magnitude greater than drift at the end of the spin up. This indicates that a 100-year biogeochemical spinup is sufficient to eliminate model drift that would impact our upper ocean analysis. Following spinup, the model was then run with NCEP/NCAR daily forcing fields for 1948-2009.

Model physics across the North Atlantic, as well as pCO_2, DIC and ALK at the Bermuda Atlantic Time Series (Bates, 2007) and in the subpolar North Atlantic have been compared to results from a previous simulation using with this same model forced with observed atmospheric pCO_2 for 1992-2006 (Ullman et al., 2009). Comparison of this simulation to estimates of the pre-industrial vertical profile of DIC in the subpolar gyre indicates good performance by the model (Supplementary Figure 1 and text). Mikaloff-Fletcher et al. (2007) estimated the pre-industrial, or 'natural', air-to-sea CO_2 flux in the North Atlantic with an ocean inversion that incorporated climatological circulations estimated from 10 ocean circulation models. For the North Atlantic from 0° to 75°N, they find an uptake of 0.27±0.07 PgC/yr. The mean natural CO_2 flux averaged over the same spatial domain in our simulation is consistent, 0.23 PgC/yr. In total, our comparison to available data indicate that the model is capable of robustly simulating the carbon biogeochemistry of the North Atlantic and its response to climate variability.

2.2 Post-processing

CO_2 flux into the ocean is proportional to the partial pressure difference between the atmosphere and ocean surface: $\Delta pCO_2 = pCO_2^{atm} - pCO_2^{ocn}$. In this analysis, we can directly relate higher pCO_2^{ocn} to a reduction in CO_2 flux, since atmospheric pCO_2 is fixed. ΔpCO_2 variability sets the sign and magnitude of flux changes on both seasonal and interannual timescales (Takahashi et al. 2009, Watson et al. 2009, LeQuéré et al. 2010). pCO_2 is decomposed into contributions from temperature and chemical effects using model output and the full

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carbonate equations (Follows et al., 2006). As in Ullman et al. (2009), pCO_2-SST is found by allowing only SST to vary in the full carbonate equations for pCO_2, i.e. all other variables (DIC, ALK, SSS, phosphate, silica) are held constant at their long-term mean values; pCO_2-chem is found by holding SST constant and allowing the rest of the input variables to vary; for pCO_2-DIC, only DIC varies.

Model diagnostics for DIC are the monthly mean tendency terms (in mmol/m^3/yr) due to individual modeled processes and are calculated at each time step during the model simulation (Ullman et al., 2009). Monthly mean diagnostics for the surface layer DIC change due to horizontal and vertical advection and diffusion, net biological processes (primary production and respiration), freshwater input/removal, and air-sea CO_2 flux are used.

The AMO index for the model is calculated using modeled SST and observed global Had1SSTv1.0 (Rayner et al., 2003) using the approach of Wang and Dong (2010). This approach regresses the area-weighted global mean Had1SST time series onto area-weighted basin-wide mean North Atlantic SST time series (NASST). This regressed index is subtracted from the total NASST to define the AMO. The combined SST signal is, thus, decomposed into contributions from globally increasing SST (SST trend) and the internal variability of the AMO (Figure 1d). In order to focus on the decadal timescale variability, all timeseries are smoothed with a standardized 121-month box smoother.

3 Results

3.1 Multidecadal Variability

To determine the leading mode of variability in surface ocean pCO_2, principle component analysis is employed. The first empirical orthogonal function (EOF1) patterns and smoothed principle components (PCs) for monthly, 13-month smoothed total pCO_2 and the SST contribution to pCO_2 (pCO_2-SST) are shown in Figure 1a-c. To determine the change in pCO_2 anomalies described by EOF1 at a specific point in time, the value of the PC1 at that time can be multiplied by the EOF1 pattern. The percent of variance in the total field explained by the EOF1 pattern is 18% and 38% for pCO_2 and pCO_2-SST, respectively. In both cases, the EOF1 patterns are statistically distinct from their EOF2 patterns, which are discussed in section 4. This EOF analysis unveils the basin-scale coherent variability. There is remaining variability in coherent secondary large-scale modes (e.g. EOF2) or at scales smaller than the whole basin. That large-scale modes of climatic variability tend to capture 10-40% of variance has been documented

across many climate variables, including global SST and tropospheric winds (von Storch and Zwiers, 1999), Southern Ocean geopotential heights (Thomson and Wallace, 2000), and pCO_2 throughout the Pacific (McKinley et al. 2004, 2006). That pCO_2 EOF1 captures the patterns of multi-decadal large-scale change is further evidenced by plots of 20-year anomalies of pCO_2 , (Figure S2).

The correlation between PC1-pCO_2 and the area-weighted basin-averaged SST is 0.88 (Figure 1c, Table S1). An increase in temperature increases pCO_2 by reducing solubility, which is illustrated by the pCO_2-SST EOF1 pattern. PC1- pCO_2 and PC1- pCO_2-SST are highly correlated (Figure 1c, r = 0.91), but have distinct EOF1 patterns, particularly in the subpolar gyre (Figure 1a,b). This is consistent with the pCO_2 in the subpolar gyre also being significantly impacted by changes in DIC supply which in turn, are associated with the AMO. EOF1 for pCO_2-chem and pCO_2-DIC explain 32% and 25% of the variance, respectively (Figure 2a,b), and these PC1's are highly correlated with the AMO, r = 0.99, 0.96, respectively (Figure 2d, Table S1).

Alkalinity can also affect pCO_2-chem since increased alkalinity reduces pCO_2. PC1 for EOF1 of pCO_2-ALK (Figure 2c) does not correlate highly to PC1's of total pCO_2 or pCO_2-chem (r=-0.25; r=0.44, respectively), or to the AMO (see supplementary table 1). Though alkalinity does contribute to the spatial pattern shown in the EOF1 of pCO_2-chem, the temporal evolution of this pattern differs substantially and is not strongly connected to the AMO or to EOF1 of pCO_2. Therefore, we focus on the more direct relationship between pCO_2-DIC and pCO_2-chem for the rest of the paper, and reserve the alkalinity relationships for future in-depth analysis.

The AMO, an index of internal North Atlantic SST variability, declines (cools) until 1975 and rises thereafter (Figure 1d). Taking the last half of the timeseries as an example, increasingly positive AMO corresponds to a decrease in pCO_2-chem, with the strongest declines in the subpolar gyre and driven by reduced pCO_2-DIC (Figure 2). This occurs in opposition to the direct effect on pCO_2 of warmer NASST (Figure 1b,c), driven jointly by the increasingly positive AMO and the warming trend (Figure 1d). SST and chemical terms vary inversely because higher SST enhances stratification, leading to a shoaling of mixed layer depths over most of the gyre (Figure S2). This shoaling in turn limits the amount of deep, carbon-rich water that is mixed to the surface, reducing pCO_2-DIC and pCO_2-chem (Ullman et al., 2009). The correlation of PC1-

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pCO_2-chem and PC1-pCO_2-DIC with PC1-pCO_2-SST are 0.90 and 0.91, respectively (Table S1). Mechanisms of AMO impacts on pCO_2-chem in the subpolar gyre will be explored further below.

3.2 Regression Analysis

Regression of the AMO, SST trend, and total SST (Figure 1d) onto monthly pCO_2, pCO_2-SST and pCO_2-chem further illustrates that temperature and chemical responses tend to act in opposition to one another, damping total pCO_2 responses across the basin (Figure 3). This analysis compliments the above EOF analysis by allowing the use of the same index of temporal variability across all fields. Previous studies with observations and models have shown that the pCO_2-chem dominates the seasonality of pCO_2 in the subpolar gyre, via strong vertical supply of DIC in winter that drives up pCO_2 and biological DIC drawdown in summer that drives pCO_2 down. Temperature impacts oppose these seasonal oscillations, but are of weaker amplitude (Kortzinger et al. 2008; Takahashi et al. 2002). Models have shown similar opposing influences with respect to interannual variability (Ullman et al. 2009; McKinley et al. 2004). These regressions illustrate that positive AMO leads to higher pCO_2-SST throughout the basin (Figure 3b). The response is strongest north of 35°N with a clear maximum to the east of Newfoundland. Simultaneously, positive AMO is associated with a reduction in pCO_2-chem (Figure 3c). The pCO_2-chem signal is also strongest to the north. The overall effect is a decrease in total pCO_2 north of 45°N and a slight increase in pCO_2 in the eastern subtropical gyre (Figure 3a).

When responding to the global SST trend, pCO_2-SST more heavily controls the response of the total pCO_2 field (Figure 3d,e). The pCO_2-SST response is strongest along the Gulf Stream and east of Newfoundland, and also increases somewhat off the coast of Europe and Africa. pCO_2-chem exhibits some decline in the Gulf Stream region, and has a small response elsewhere (Figure 3f).

Regression with the total NASST timeseries (Figure 1) illustrates the combined effects of the AMO and trend signals (Figure 3g-i). A positive anomaly of NASST depresses total pCO_2 in the subpolar gyre, consistent with the AMO impact found above. Positive NASST also increases total pCO_2 off North Africa, consistent with the impact of the SST trend. pCO_2-SST increases both off Africa and has a strong maximum in the Gulf Stream region east of Newfoundland with positive NASST anomlies. The pCO_2-chem response is slightly weaker in the subpolar gyre than for the AMO alone (Figure 3a,i).

3.3 DIC Diagnostics

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To further investigate the chemical term response to the AMO, model diagnostics for the DIC field are regressed upon the AMO index. Diagnostics are modeled rates of change in DIC due to one of five processes that have been saved at every model time-step. Physical processes are separated into horizontal advection and diffusion (DIC-horz), and vertical advection and diffusion (DIC-vert). DIC-phys is the sum of vertical and horizontal transport, showing the net effect of physical transport on DIC (Figure 4). The rate of DIC supply is also affected by biological processes involving DIC incorporation into organic matter and remineralization back to inorganic (DIC-bio), net precipitation/evaporation that dilutes or concentrates DIC (DIC-fresh) and the air-sea flux of CO_2 (DIC-flx) (Figure 5). The focus on DIC is justified by the fact that pCO_2-chem change has the same pattern and is highly correlated with pCO_2-DIC change (Figure 2). The focus on the AMO is justified by its strong imprint on pCO_2 through pCO_2-chem (Figure 2, 3).

For the long-term average, vertical advection and diffusion are positive along the Gulf Stream and in the subpolar gyre due to deep winter mixed layer depths (MLD) that mix up high-DIC water from below (Figure 4a). Horizontal DIC advection and mixing removes this vertically supplied DIC along the Gulf Stream and in the western subpolar gyre (Figure 4b). While the vertical and horizontal components tend to have opposing influences, the net effect is a positive DIC supply to the subpolar gyre, as shown by mean DIC-phys (Figure 4c). With positive AMO, vertical advective and diffusive fluxes of DIC decrease in the Irminger Sea and Iceland basin, while they increase in the Labrador Sea and east of Newfoundland (Figure 4d). These changes are consistent with AMO-related MLD changes outside of the Labrador Sea (Figure 5) and change in the basin-scale barotropic streamfunction indicating a weakened subpolar gyre (Figure 6). The effect of this is to shift the central DIC-vert maximum to the west. With positive AMO, horizontal advection and diffusion largely respond to changes in vertical advection and diffusion, with less horizontal divergence (a positive change) in regions where the vertical supply is reduced (Figure 4e). The net effect shown by DIC-phys reveals an overall reduction in DIC supply (Figure 4f), consistent with a weaker subpolar gyre circulation and shallower MLDs that reduce the vertical supply of DIC. Hakkinen and Rhines (2009) illustrate and increased penetration of subtropical waters into the subpolar region from the 1990s to the 2000s, consistent with a weaker subpolar gyre circulation. The changes in MLD and streamfunction are also in agreement with results from Zhang (2008) who links the observed spindown of the subpolar gyre

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in the 1990's to an enhanced MOC using a combination of satellite altimeter observations and results from a 1000-year coupled ocean-atmosphere model simulation.

Mean DIC impacts from physics, biological processes, freshwater and air-sea flux are shown in Figure 7a-e. The net impact of biology is to remove DIC from the surface of most of the region, with the most intense removal along the Gulf Stream (Figure 7a). The smaller impact of evaporation and precipitation is to concentrate DIC in the subtropics and to dilute it in the subpolar gyre (Figure 7b). The air-sea CO_2 flux term is also small, positive north of about 35°N and negative to the south (Figure 7c). AMO-related change in the biological removal of DIC indicates additional removal (negative anomaly) occurring in the same region where horizontal flux increases, consistent with biological stimulation through an increased supply nutrients from the subtropical subsurface along the "nutrient stream" (Williams et al., 2006). There is reduced biological productivity, and thus a reduction of DIC loss (a positive DIC anomaly), in other parts of the basin that are consistent with satellite observations from the late 1990s to the mid-2000s (Behrenfeld et al. 2006). Changes in surface ocean DIC content due to freshwater fluxes and air-sea CO_2 flux with the AMO are small. Across the basin, the net DIC change associated with AMO is negative, with the strongest negative changes occurring in the subpolar gyre (Figure 2b, 3c)

4 Discussion and Conclusions

In this North Atlantic regional model forced with pre-industrial pCO_2 and realistic climate from 1948-2009, SST is the dominant driver of pCO_2 variability, with both long-term anthropogenic warming and the AMO playing important roles. The AMO strongly influences chemical change, which in turn is mostly driven by DIC. DIC changes, in turn, are due primarily to changes in vertical and horizontal advection and mixing. Changing biology has the most important secondary effect, and largely damps the anomalies caused by advection and mixing. Freshwater and CO_2 fluxes changes are slight.

Our findings linking the AMO to natural carbon cycle variability in the North Atlantic are consistent with the study of Séférian et al. (2013) who also found an AMO-like signal dominated North Atlantic pCO_2 variability in a 1000-year Earth System Model simulation with constant pCO_2. Other studies have focused on the relationship between the North Atlantic Oscillation (NAO) and CO_2 flux using models and observations (Loptien and Eden 2010; Ullman et al. 2009; Schuster et al., 2009; Thomas et al., 2008). Consistent with these previous studies, the

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NAO is the second mode of variability in this simulation (Figure S4, S5), and the corresponding principle components are highly correlated with the NAO (Table S2). The shorter timeframe for most previous studies explains, in part, the difference in attribution to the AMO as opposed to NAO. Our results are broadly consistent with previous studies in the finding that physical variability is the dominant driver of variability in the North Atlantic surface ocean carbon cycle.

The NAO and AMO may, in fact, be linked through the Meridional Overturning Circulation (MOC), with a positive NAO enhancing MOC, which over time warms SSTs and leads to a positive AMO. The precise mechanisms remain in debate due to different model findings and a lack of observational constraints (Delworth & Mann, 2000; Knight et al., 2005; Dima & Lohmann, 2007; Latif et al., 2006). In this simulation, the NAO and MOC are significantly correlated (r = 0.57, Table S1) and there is also a high correlation (r = 0.86) between the NAO (Figure S3) and the 15-year lagged AMO. These correlations are consistent with the above-postulated NAO-MOC-AMO relationship. On the other hand, Booth et al. (2012) suggest that the AMO may be driven, in fact, by atmospheric aerosol variability, so it is possible that there is no direct AMO-MOC relationship. Future modeling and observations should further elucidate these connections.

We find multidecadal variability in the natural carbon cycle of the surface North Atlantic to be dominated by the SST trend and multidecadal SST variation captured by the AMO index. Variability linked to AMO influences both pCO_2-SST and pCO_2-chem. In the subpolar gyre, the positive SST influence on pCO_2 is overwhelmed by reduced supply of DIC to the surface ocean through mixing and advection, the net impact being reduced pCO_2. The reduction in mixing is associated with shoaling of MLDs and a weaker subpolar gyre circulation, both associated with warmer SSTs (positive AMO). In the subtropics, the SST impact is stronger and thus pCO_2 is increased under the influence of positive AMO and positive SST trend.

These findings are consistent with observed relationships between trends in surface ocean pCO_2 and trends in atmospheric pCO_2 since the 1980s (Fay and McKinley, 2013). In the North Atlantic subpolar gyre, trends in surface ocean pCO_2 lagged the trend in atmospheric pCO_2 from the early to mid 1990s to the late 2000s, which is consistent with the AMO and the SST trend reducing DIC supply to the subpolar gyre as found in this study. On smaller spatial scales and shorter timeframes, trends in ocean pCO_2 can differ (Fay and McKinley, 2013; Metzl 2010, Watson et al 2009, Schuster et al. 2009), which can be reasonably attributed to shorter-term and

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smaller spatial scale variability. We also find that warming has contributed to the observed pCO_2 increase from the 1980-90s through the 2000s throughout the basin. These model results allow a mechanistic attribution of these observed changes in North Atlantic pCO_2 to the combined effect of the AMO and a positive SST trend due to anthropogenic climate change.

Acknowledgements

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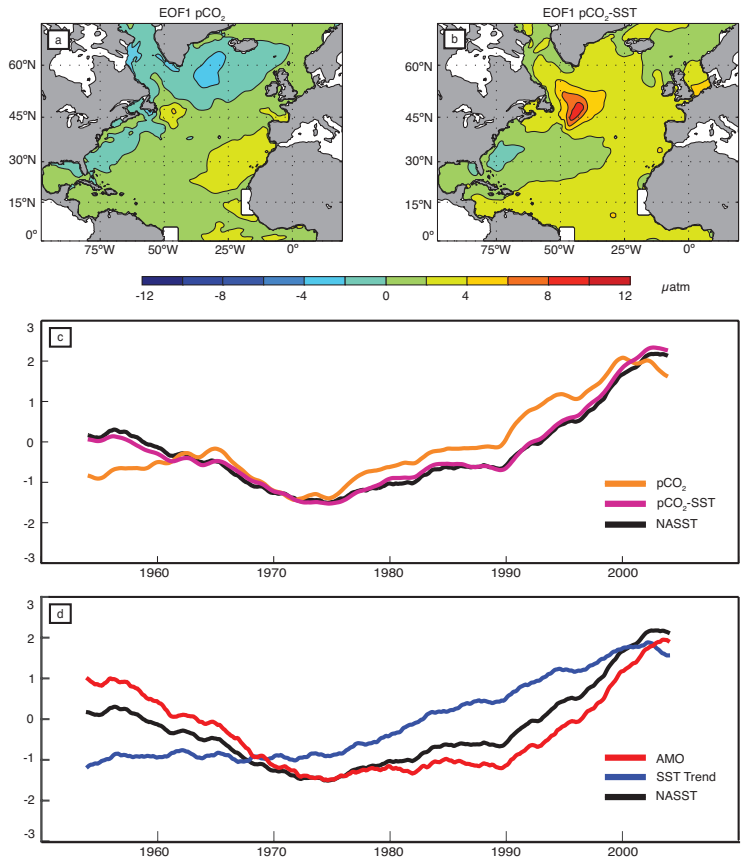

Figure 1: a) EOF1 of total pCO_2 (µatm), b) EOF1 of pCO_2-SST (µatm), explaining 18% and 38% of total variance, respectively. c) PC1- pCO_2 (orange), PC1- pCO_2-SST (pink) and area-weighted, basin-averaged standardized North Atlantic SST time series (black), d) Area-weighted, basin-averaged (0-70N, 98W-19.5E) North Atlantic SST from Had1SST (black), global area-weighted SST regressed onto North Atlantic SST (blue), and AMO index created by subtracting the global regression from the North Atlantic SST (red). All indices are standardized by 1-sigma. Timeseries smoothed with a 121-month box smoother. Two small coastal areas off Africa and South America were excluded in a) and b) due to the presence of localized, anomalously strong upwelling in the early 1960's that precluded elucidation of the large-scale pattern.

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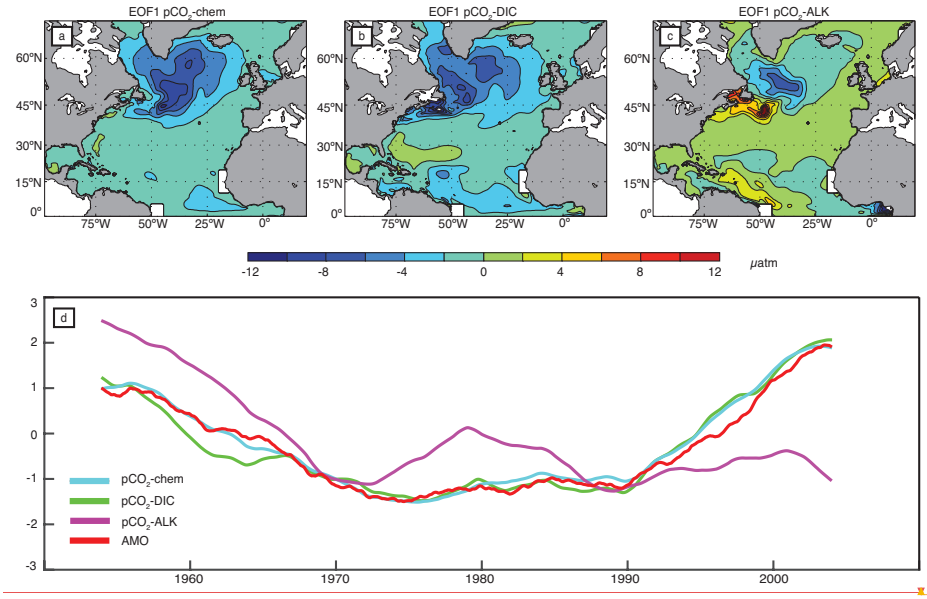

Figure 2: a) EOF1 pCO_2-chem (µatm), b) EOF1 pCO_2-DIC (µatm), c) EOF1 pCO_2-ALK explaining 32%, 25%, and 19% of total variance, respectively, d) PC1- pCO_2-chem (cyan), PC1-pCO_2-DIC (green) PC1-pCO_2-ALK (magenta) and AMO index (red), all standardized. Timeseries smoothed with a 121-month box smoother.

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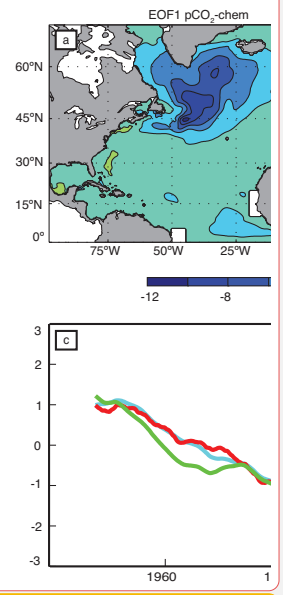

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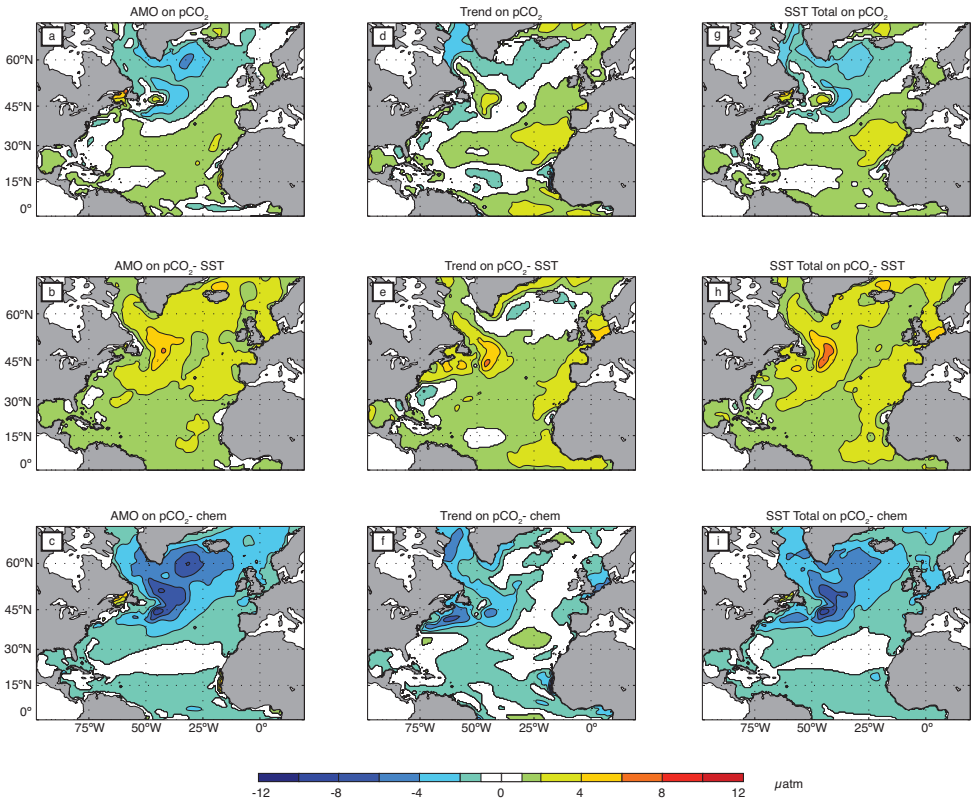

Figure 3: 121-month box-smoothed AMO regressed onto unsmoothed, monthly a) pCO_2, b) pCO_2-SST, c) pCO_2-chem. SST Trend regressed onto d) pCO_2, e) pCO_2-SST, f) pCO_2-chem. NASST (AMO + SST Trend) regressed onto g) pCO_2, h) pCO_2-SST, i) pCO_2-chem. Regressions calculated from 1953 through 2005. Values <0.5 and >-0.5 μatm are whited out to highlight regions experiencing the most substantial changes.

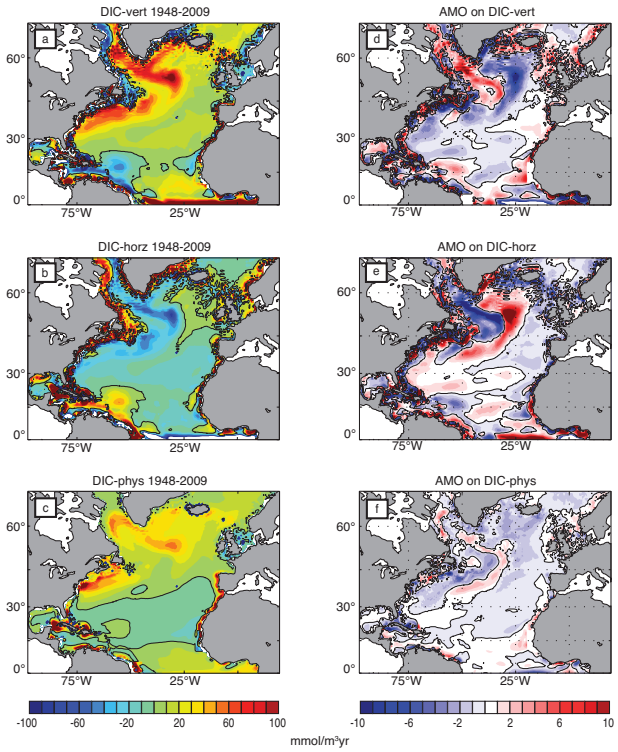

Figure 4: DIC diagnostics. Left column: 1948-2009 Mean a) DIC-vertical, b) DIC-horizontal, c) DIC-physical where DIC-physical is the sum of DIC-vertical and DIC-horizontal. Right column: AMO regressed onto d) DIC-vertical, e) DIC-horizontal, f) DIC-physical. Units mmol/ m^3/yr.

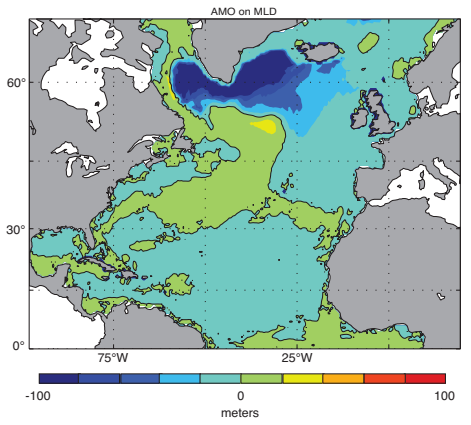

Figure 5: Regression of AMO on Mixed Layer Depth (MLD). Negative values denote a shoaling of MLD.

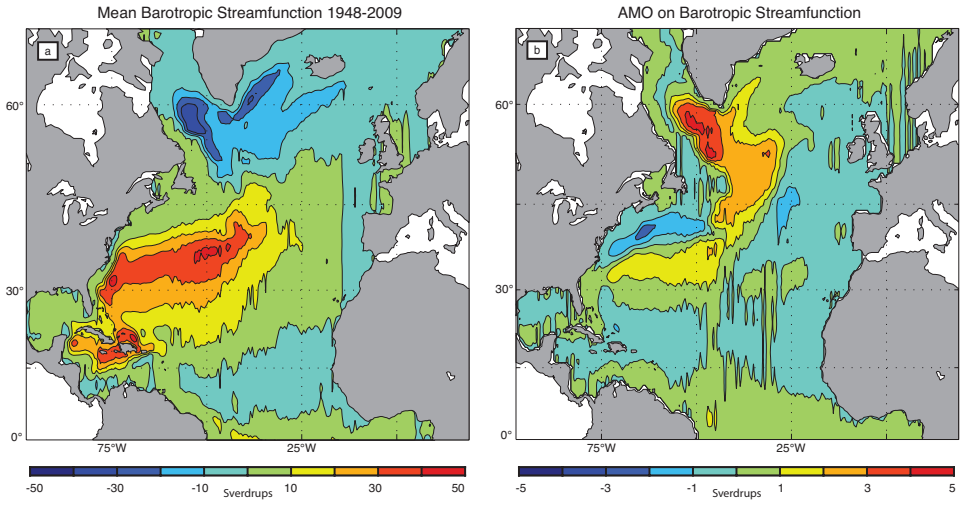

Figure 6: a) 1948-2009 mean barotropic streamfunction and b) AMO regressed onto barotropic streamfunction anomalies. Positive values denote clockwise motion. Units: Sverdrups (1 Sv = 10^6 m^3*s^{-1}).

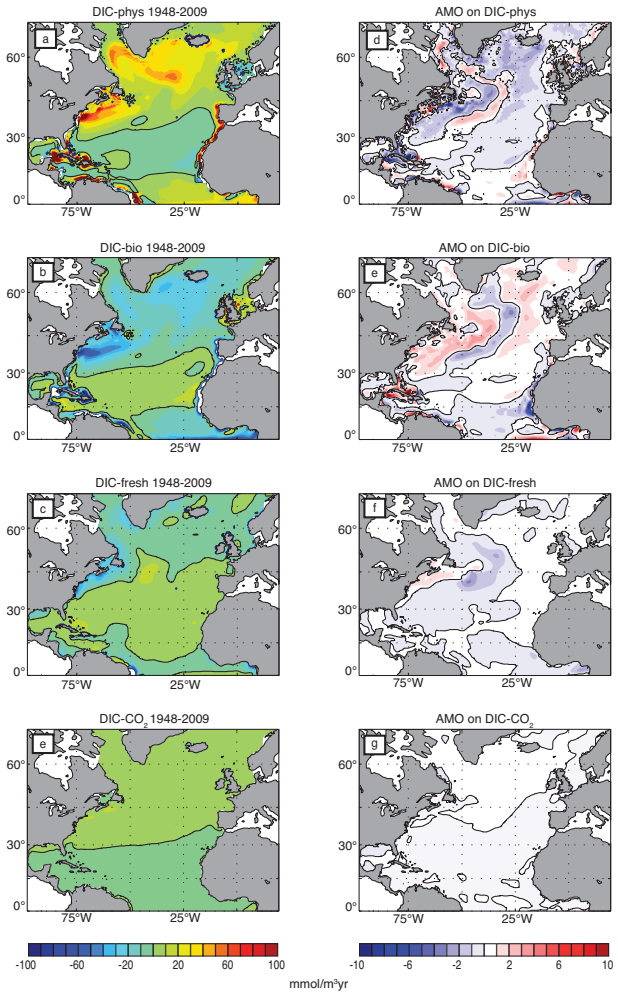

Figure 7: DIC diagnostics. Left column: 1948-2009 Mean a) DIC-physical, b) DIC-bio, c) DIC-fresh, d) DIC-CO_2flux . Right column: AMO regressed onto e) DIC-physical, f) DIC-bio , g) DIC-fresh, h) DIC-CO_2flux. Units mmol/m^3/yr

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Supplementary Material for: Climate impacts on multidecadal pCO_2 variability in the North Atlantic: 1948-2009

Melissa L. Breeden and Galen A. McKinley

Supplementary Text:

To assess the model's ability to capture the vertical distribution of DIC, we compare here the 0-4000m average 1948-2009 model DIC profile to the GLODAP (Key et al. 2004) estimate of pre-industrial DIC across the subpolar gyre (35-55N, 5-60E, Figure S1). The model falls within the uncertainty of the observed estimate from 0-2000m, and is a few percent below the observed estimate from 2000-4000m . The average annual maximum MLD of this region is 284m and the annual maximum MLD across all points in the region is 3215m. Thus, annual vertical mixing occurs predominantly in the depth range where the model captures the observed estimate. In sum, this comparison suggests that model is a reasonable tool for studying how climate variability impacts variability in the vertical supply of carbon from the deep to surface ocean in the subpolar North Atlantic.

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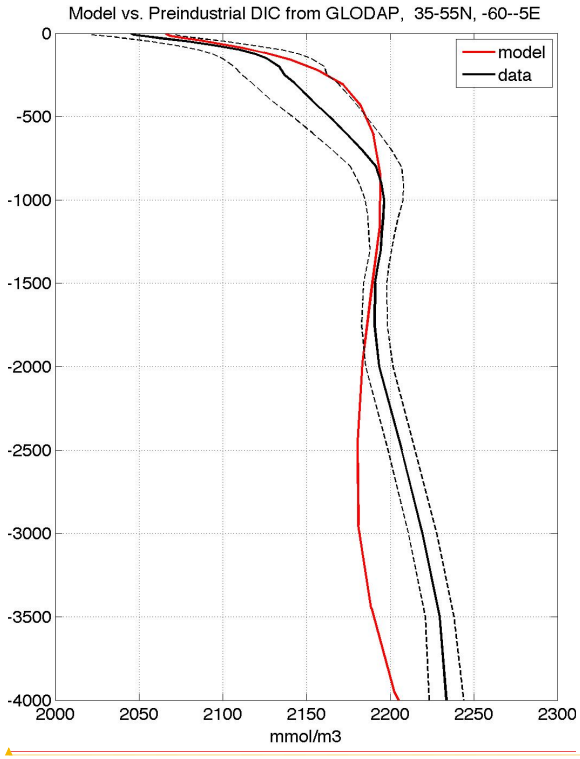

Figure S1: 35-55N, 5-60E average profile of 1948-2009 Model (red) and GLODAP preindustrial DIC (black) in mmol/m^3.

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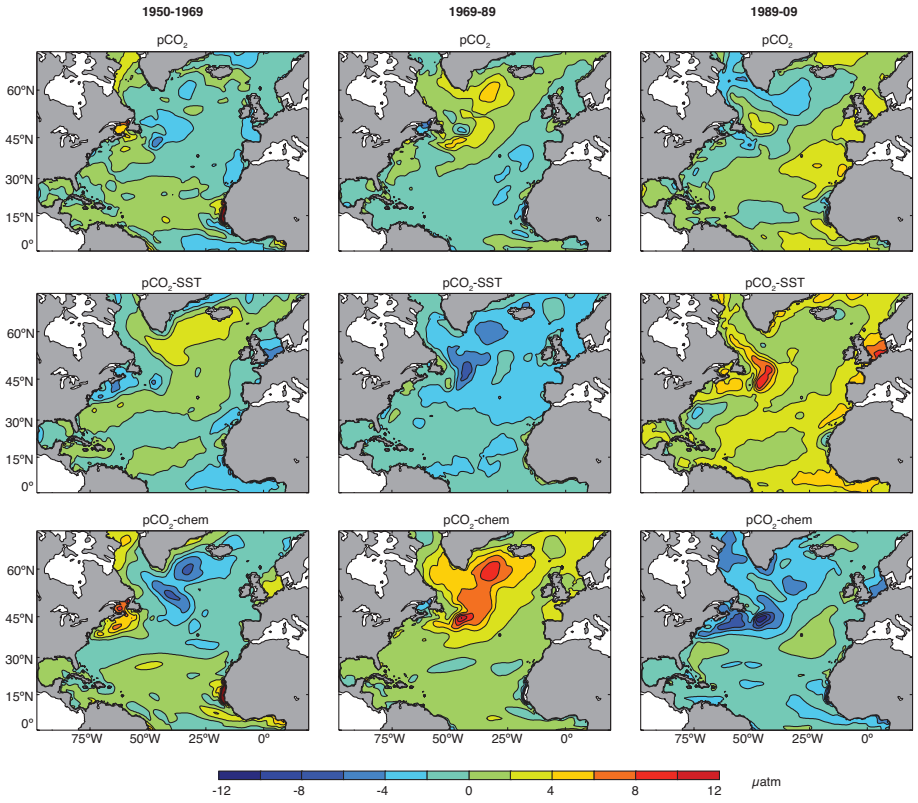

Figure S2: 20-year mean anomalies with respect to the 1948-2009 average for: a-c) pCO_2, d-f) pCO_2-SST, g-i) pCO_2-chem.

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Figure S2: Regression of AMO on Mixed Layer Mixed Layer Depth (MLD). Negative values denote a shoaling of MLD. ... [1]

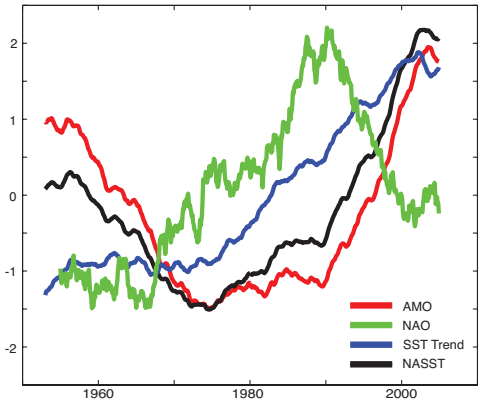

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Figure S3: Same indices as in Figure 1d, as well as 121-month smoothed, standardized NAO index from NOAA ESRL (http://www.esrl.noaa.gov/psd/data/climateindices/list/).

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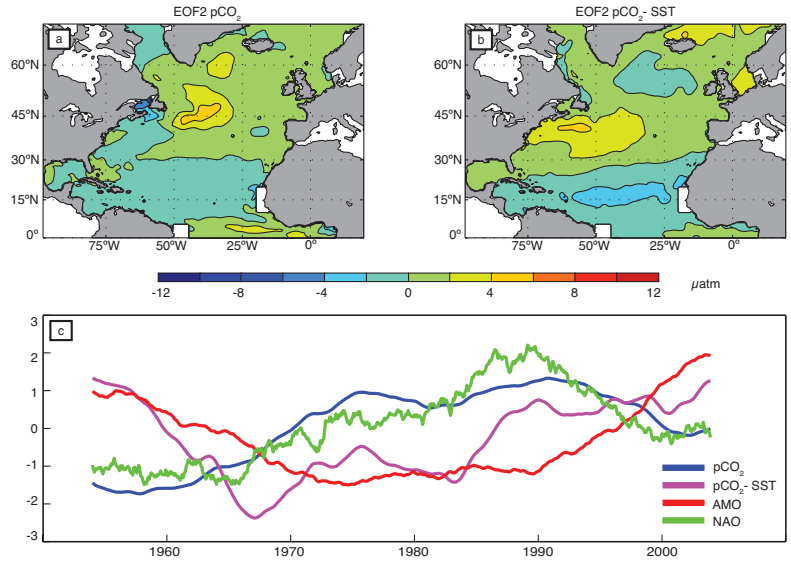

Figure S4: a) EOF2 of total pCO_2 (µatm), b) EOF2 of pCO_2-SST (µatm), explaining 13% and 12% of total variance, respectively. c) PC2- pCO_2 (blue), PC2-pCO_2-SST (pink), AMO index (red), and NAO index (green). Timeseries are smoothed with a 121-month box smoother.

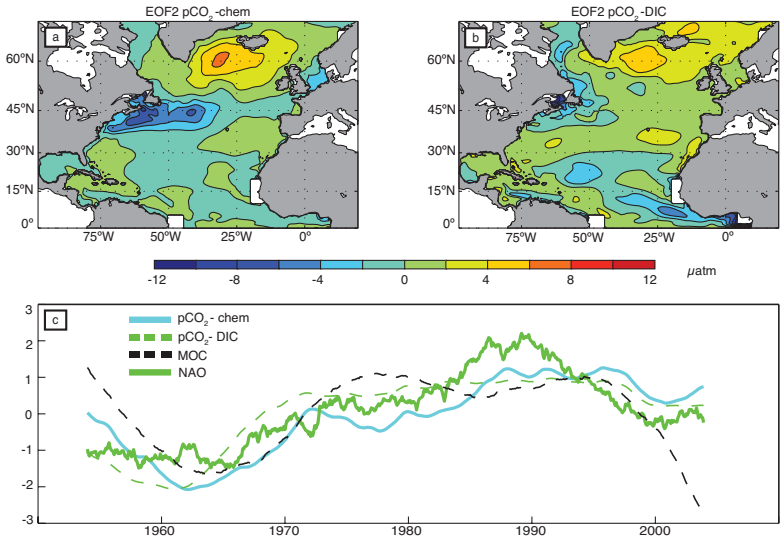

Figure S5: a) EOF2 pCO_2-chem (µatm), b) EOF2 pCO_2-DIC (µatm), each explaining 14% of total variance of each respective field. c) PC2- pCO_2-chem (cyan), PC2- pCO_2-DIC (green dash), NAO index (green), the modeled maximum Meridional Overturning Circulation at 45°N (MOC, black dash), all standardized. Timeseries are smoothed with a 121-month box smoother.

	PC1-pCO_2	PC1-pCO_2-SST	PC1-pCO_2-chem	PC1-pCO_2-DIC	PC1-pCO2-ALK	SST	AMO	SST Trend	NAO	MOC
PC1-pCO_2	**1.0**	**0.91**	**0.66**	**0.67**	**-0.25**	**0.88**	**0.61**	**0.94**	**0.23**	0.0026
PC1-pCO_2-SST		**1.0**	**0.90**	**0.91**	.05	**1.0**	**0.86**	**0.78**	-0.069	**-0.12**
PC1-pCO_2-chem			**1.0**	**0.98**	**.44**	**0.92**	**0.99**	**0.45**	**-0.42**	**-0.25**
PC1-pCO_2-DIC				**1.0**	**.35**	**0.93**	**0.96**	**0.49**	**-0.38**	**-0.18**
PC1-pCO2-ALK					**1.0**	**.12**	**.48**	**-.48**	**-.70**	**-.08**
SST						**1.0**	**0.90**	**0.74**	**-0.11**	**-0.13**
AMO							**1.0**	**0.37**	**-0.49**	**-0.32**
SST Trend								**1.0**	**0.51**	**0.21**
NAO									**1.0**	**0.57**
MOC										**1.0**

Table S1: Correlation between first principle components of the EOFs for pCO_2 and its components, climate indices, and the modeled maximum Meridional Overturning Circulation (MOC) at 45°N. Index and MOC correlations are also shown. Bold indicates significance at the 95% level.

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	PC2-pCO_2-	PC2-pCO_2-SST	PC2-pCO_2-chem	PC2-pCO_2-DIC	SST	AMO	SST Trend	NAO	MOC
PC2-pCO_2-	**1.0**	0.080	**0.80**	**0.96**	**-0.18**	**-0.59**	**0.50**	**0.89**	**0.56**
PC2-pCO_2-SST		**1.0**	**0.52**	-0.039	**0.70**	**0.60**	**0.57**	**0.18**	**0.37**
PC2-pCO_2-chem			**1.0**	**0.82**	**0.28**	**-0.11**	**0.75**	**0.82**	**0.65**
PC2-pCO_2-DIC				**1.0**	**-0.12**	**-0.51**	**0.52**	**0.83**	**0.51**

Table S2: Correlation between second principle components of the EOFs for pCO_2 and its components, climate indices, and MOC. Bold indicates significance at the 95% level.