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Key Points:
- Observational evidence shows that the mechanisms underlying seasonal variability in $\Delta p\mathrm{CO}_2$ are not the same as those underlying interannual variability
- The presence of a vigorous spring bloom and the resultant phytoplankton succession dominate seasonal $\Delta p\mathrm{CO}_2$ in subpolar waters
- Long-term observations of ocean CO_2 are required to distinguish seasonal and interannual controls on $\Delta p\mathrm{CO}_2$

Supporting Information:
- Supporting Information S1

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Controls on Open-Ocean North Atlantic $\Delta p\mathrm{CO}_2$ at Seasonal and Interannual Time Scales Are Different

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Abstract The North Atlantic is a substantial sink for anthropogenic CO_2. Understanding the mechanisms driving the sink's variability is key to assessing its current state and predicting its potential response to global climate change. Here we apply a time series decomposition technique to satellite and in situ data to examine separately the factors (both biological and nonbiological) that affect the sea-air CO_2 difference ($\Delta p\mathrm{CO}_2$) on seasonal and interannual time scales. We demonstrate that on seasonal time scales, the subpolar North Atlantic $\Delta p\mathrm{CO}_2$ signal is predominantly correlated with biological processes, whereas seawater temperature dominates in the subtropics. However, the same factors do not necessarily control $\Delta p\mathrm{CO}_2$ on interannual time scales. Our results imply that the mechanisms driving seasonal variability in $\Delta p\mathrm{CO}_2$ cannot necessarily be extrapolated to predict how $\Delta p\mathrm{CO}_2$, and thus the North Atlantic CO_2 sink, may respond to increases in anthropogenic CO_2 over longer time scales.

Plain Language Summary As atmospheric carbon dioxide (CO_2) concentrations rise due to anthropogenic emissions, the ocean is taking up more CO_2, a process known as the oceanic CO_2 sink. The North Atlantic is a major anthropogenic CO_2 sink; however, factors that drive variability in the sink are still under investigation. In order to assess the sink's current state and future with continued climate change, we need to understand what affects the North Atlantic CO_2 sink. Often, the factors that affect oceanic uptake of CO_2 are explored on a seasonal time scale. Here we take a longer view, examining the factors that may affect ocean uptake on interannual time scales. We find that the factors are different, depending on whether we assess the short or long term. In building models of ocean response to future climate change, we cannot extrapolate the response of ocean CO_2 uptake to seasonal variability out to longer time scales.

1. Introduction

On multidecadal time scales, the ocean is a key route for removal of anthropogenic CO_2 from the atmosphere, taking up approximately one third of emissions since preindustrial times (Khatiwala et al., 2013). The high-latitude North Atlantic has one of the highest uptake rates of atmospheric CO_2 per square meter (Mikaloff-Fletcher et al., 2006), accounting for 23% of oceanic anthropogenic CO_2 storage, while only constituting 15% of the global ocean surface area (Sabine et al., 2004). However, recent studies suggest that the North Atlantic CO_2 sink may be weakening by up to 50% in the southeastern subpolar gyre (Schuster et al., 2009). Whether the North Atlantic is a source or sink of atmospheric CO_2 varies both spatially and temporally due to the interacting effects of seawater temperature, ocean circulation, and biological activity (Watson et al., 2009).

During air-sea gas exchange the CO_2 concentration difference across the boundary layer determines the net direction of CO_2 transfer (Woolf et al., 2016), that is, the difference between the partial pressure of CO_2 ($p\mathrm{CO}_2$) in seawater and the overlying atmosphere ($\Delta p\mathrm{CO}_2$). This approach ignores the impact of turbulent exchange and vertical temperature gradients near the sea surface but provides a useful broad-scale indicator of the direction of CO_2 transfer. Where $\Delta p\mathrm{CO}_2$ is positive (seawater $p\mathrm{CO}_2 >$ atmospheric $p\mathrm{CO}_2$), the water is oversaturated, implying a net flux from sea to air, that is, a potential CO_2 "source." The opposite case, where $\Delta p\mathrm{CO}_2$ is negative and the ocean is undersaturated, implies a CO_2 "sink." Atmospheric $p\mathrm{CO}_2$ is homogeneous relative to seawater, so seawater $p\mathrm{CO}_2$ is typically the dominant control on $\Delta p\mathrm{CO}_2$ direction. Thus, biogeochemical and hydrographic processes can modify $\Delta p\mathrm{CO}_2$ if they alter the seawater $p\mathrm{CO}_2$. Cooler water has a greater capacity to store dissolved inorganic carbon (DIC) than warm water, as CO_2 solubility is inversely proportional to water temperature. Cooler water reduces seawater pCO_2, helping to drive negative ΔpCO_2, while warming has the opposite effect. Net community production (NCP, primary production minus respiration) takes up DIC from the seawater through photosynthesis, decreasing seawater pCO_2 and contributing to negative ΔpCO_2. Calcification consumes DIC but is a CO_2 source due to the accompanying net release of CO_2 into the water (Frankignoulle et al., 1994), which may have a significant localized impact in the North Atlantic (Shutler et al., 2013). The net effect of the combination of physical and biological drivers results in an overall CO_2 sink in the subpolar North Atlantic and a neutral to weak sink in the subtropical North Atlantic (Schuster et al., 2013).

An additional biological influence on the air-sea CO_2 flux has been posited: Phytoplankton community structure is expected to have a dominant effect (Hilligsoe et al., 2011), with functional types such as diatoms thought to export organic carbon most efficiently (Michaels & Silver, 1988). However, small phytoplankton have also been found to influence CO_2 uptake and export (Palevsky et al., 2013; Richardson & Jackson, 2007), and in the North Atlantic dinoflagellate abundance was found to strongly correlate with organic carbon flux at 2,000 m (Henson et al., 2012). While calcifying phytoplankton (e.g., coccolithophores) can also modify seawater pCO_2 during formation, they may also contribute to efficient organic carbon transfer to depth (Klaas & Archer, 2002).

The potential controls on the North Atlantic CO_2 sink at different time scales are not well understood. For example, in a model study Bennington et al. (2009) found that biological activity dominated the seasonal cycle of seawater pCO_2, but not its interannual variability. On these longer time scales, the North Atlantic Oscillation (NAO), the dominant climate variability mode in the region, could affect oceanic CO_2 uptake (Gruber et al., 2009) and interior CO_2 storage (Humphreys et al., 2016). In a positive NAO phase, the North Atlantic Current increases in strength (Visbeck et al., 2003), bringing warm waters with relatively low DIC concentration into the subpolar northeast Atlantic. Despite the warm water, the low DIC results in an intensified CO_2 sink in that region, while in the northwest Atlantic an intensified Labrador Current brings cooler waters with relatively high DIC from the Arctic, which, despite the cool water, results in a weaker CO_2 sink (Völker et al., 2002). In the subtropical Atlantic a positive NAO phase has the effect of reducing mixing and increasing surface water temperatures, which result in lower carbon uptake (Gruber, 2009).

Using observational data sets, we examine the hypothesis, suggested by a previous model study (Bennington et al., 2009), that the dominant influences on ΔpCO_2 in the North Atlantic are different at seasonal and interannual time scales. One approach is to separate the effects using a climatological mass balance technique (Ayers & Lozier, 2012); however, to specifically test the importance of potential mechanisms at different time scales, a method to decompose a time series into its seasonal and interannual components is needed. Here we apply a novel decomposition approach to a combination of satellite and in situ observations. We test whether proposed mechanisms for controlling ΔpCO_2 are potentially valid on both seasonal and longer time scales, for example, that biological effects dominate over temperature effects at high latitudes.

2. Methods

Monthly gridded fugacity of seawater CO_2 (fCO_2) for the North Atlantic was downloaded from the Surface Ocean CO_2 Atlas (SOCAT) v3 database (Bakker et al., 2016; www.socat.info) and reanalyzed to a common sea surface temperature (SST) data set (Reynolds et al., 2007) at 0.5-m depth, following the method of Goddijn-Murphy et al. (2015). Ocean pCO_2 was calculated from fCO_2 using the equations and constants provided in the seacarb R package v3 (Lavigne et al., 2011). The data were then gridded to a 1 × 1° grid following the SOCAT method (Sabine et al., 2013). To calculate the ΔpCO_2, atmospheric molar CO_2 concentration was obtained from the NOAA Marine Boundary Layer reference data set (https://www.esrl.noaa.gov/gmd/ccgg/mbl/index.html). These were converted to pCO_2(air) using the formulation of Weiss (1974), as implemented in Shutler et al. (2016). Auxiliary data sets for sea surface salinity and sea level pressure were taken from the World Ocean Atlas 2013 climatology (Zweng et al., 2013) and the National Centers for Environmental Prediction/National Center for Atmospheric Research Reanalysis data set (Kalnay et al., 1996; http://www.esrl.noaa.gov/psd/), respectively. Moderate Resolution Imaging Spectroradiometer

(MODIS)-Aqua chlorophyll concentration, photosynthetically available radiation, particulate inorganic carbon (PIC), and nighttime 11-μm (thermal band) SST data at monthly, 9-km resolution were downloaded from https://oceancolor.gsfc.nasa.gov/. The MODIS SST data were combined with chlorophyll concentration and photosynthetically available radiation data to estimate primary production using the Vertically Generalized Productivity Model (VGPM; Behrenfeld & Falkowski, 1997). PIC data were corrected for sensor saturation effects by filling erroneously missing data (Land et al., 2018). Monthly satellite-derived NCP estimates were taken from Tilstone et al. (2015). All satellite-derived data were regridded onto a 1 × 1° grid to match the resolution of the $\Delta p\text{CO}_2$ data. The NAO index used here is the monthly, principal component-based index downloaded from https://climatedataguide.ucar.edu. Monthly mean total diatom and dinoflagellate abundance data were taken from the Continuous Plankton Recorder survey (Richardson et al., 2006; data set doi: 10.7487/2016.194.1.988). MODIS data were available for the time period July 2002 to December 2014, which is used for all analyses with the exception of NCP data that end in 2010. We repeated the analysis for three alternative PP algorithms (Carr, 2001; Marra et al., 2003; Westberry et al., 2008) and two alternative NCP algorithms (Li & Cassar, 2016; Siegel et al., 2014) to investigate the sensitivity of our results to the choice of satellite PP and NCP algorithms (supporting information). For PP, the three chlorophyll-based algorithms (VGPM, Marra, and Carr) all agree on the pattern of correlation with $\Delta p\text{CO}_2$ at both seasonal and interannual time scales. Although the carbon-based productivity model (CbPM; Westberry et al., 2008) displays negative correlation between PP and $\Delta p\text{CO}_2$ in the subpolar region at seasonal time scales (consistent with other algorithms tested), in the subtropics and at interannual time scales CbPM-PP is positively correlated with $\Delta p\text{CO}_2$ (differing from other algorithms tested). For NCP at the seasonal time scale, the Li and Cassar (2016) model agrees with the Tilstone et al. (2015) results, while the Siegel et al. (2014) model shows positive, rather than negative, correlations in two midlatitude provinces. At the interannual time scale, both the Li and Cassar (2016) and Siegel et al. (2014) algorithms have several regions where NCP and $\Delta p\text{CO}_2$ are not significantly correlated. Where they are, the sign of the correlation is not necessarily the same as for Tilstone et al. (2015). Although results from different satellite algorithms are not always consistent, these are the only PP and NCP estimates available at the basin-scale and multiyear time scale that are essential for our analysis.

To overcome limitations in the spatial coverage of the SOCAT database observations from 2002 to 2014, $\Delta p\text{CO}_2$ was averaged within Longhurst provinces (Longhurst, 1998), as were all satellite-derived and Continuous Plankton Recorder data. Provinces in which $>65\%$ of the $\Delta p\text{CO}_2$ time series had missing data were excluded, as were those encompassing shelf regions. In the remaining regions, any missing province-mean monthly $\Delta p\text{CO}_2$ data (which occurred in winter in the highest-latitude provinces) were filled with climatological mean values for that region and month. On average, provinces contained 21 valid data points per month. Winter months were least well sampled, although all regions had at least 3 years of data in every month (Figure S3). To avoid spurious results, regions in which PIC is typically very low (where coccolithophore blooms are not thought to form; Moore et al., 2012) were excluded from PIC analysis (North Atlantic Subtropical Gyre, West and East).

Takahashi et al. (1993, 2002) detail a method to separate the seasonal $p\text{CO}_2$ change into temperature-driven and nontemperature-driven effects. The nontemperature-driven term is characterized as the “net biology” effect (Takahashi et al., 2002), which includes net PP, net alkalinity change due to nutrient utilization, change in surface ocean freshwater balance and carbonate production by calcifying organisms, air-sea exchange of CO_2, and change in CO_2 and alkalinity due to vertical mixing of subsurface waters. Although the nontemperature effects are dominated by biological activity, this approach is not able to distinguish the type of biological effect, for example, due to community metabolism or calcification. Therefore, to identify potential dominant biological effects, we also analyze all data following the X-11 methodology, which separates time series into seasonal, interannual, and residual components. The X-11 method was developed as an econometric tool (Shiskin et al., 1967) and has since been adapted for application to environmental time series. Here we follow the methodology of Pezzulli et al. (2005) as described in Vantrepotte and Melin (2011). A key advantage of the X-11 approach is that it permits the shape and phase of the seasonal cycle to vary from year to year; thus, the interannual component is considered more representative of the true long-term change in the time series. The time series decomposition was performed on monthly time series of the NAO index, plus $\Delta p\text{CO}_2$, SST, PP, NCP, PIC, and diatom and dinoflagellate abundance spatially averaged over Longhurst provinces.

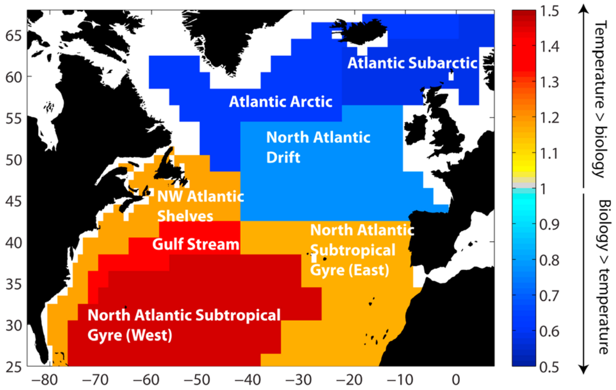

Figure 1. Relative importance of temperature and net biological effects on climatological $\Delta p\mathrm{CO}_2$ based on the decomposition of Takahashi et al. (2002). Provinces are defined and named as per Longhurst (1998).

An example of the decomposition generated by the X-11 method is given in Figure S4. The raw time series of $\Delta p\mathrm{CO}_2$, SST, and PP in the Atlantic Subarctic province are dominated by seasonal variability, making subtle changes to the phasing of the seasonal cycle, or any interannual variability, difficult to discern. The decomposition of the time series into interannual, seasonal, and residual components clarifies the picture; the interannual component shows that PP declined between 2004 and 2006 but thereafter had an increasing trend to 2012. In the following, we calculate the nonparametric Spearman correlation coefficient between the different components of $\Delta p\mathrm{CO}_2$ and SST, PP, NCP, PIC, diatom and dinoflagellate abundance, and the NAO index. Due to the substantial temporal autocorrelation in all time series, the calculated correlation coefficient is likely to be inflated. Therefore, we do not use the correlation results to test specific hypotheses (in a statistical sense) but rather to identify the spatial patterns of positive or negative correlation. We also acknowledge explicitly that correlation does not necessarily equal causation. We also calculate nonparametric partial correlation coefficients to assess association between $\Delta p\mathrm{CO}_2$ components and potential drivers while controlling for the effect of other variables (e.g., Brown & Hendrix, 2014). For example, we wish to investigate whether the seasonal component of $\Delta p\mathrm{CO}_2$ is correlated with PIC; however, PIC is also potentially correlated with NCP, which is itself correlated with $\Delta p\mathrm{CO}_2$. Partial correlation analysis allows us to determine whether PIC is statistically significantly correlated with $\Delta p\mathrm{CO}_2$ while controlling for the effect of NCP. We also test the correlation between $\Delta p\mathrm{CO}_2$ and NCP, and between $\Delta p\mathrm{CO}_2$ and NAO while controlling for SST, and the correlation between $\Delta p\mathrm{CO}_2$ and dinoflagellate abundance while controlling for diatom abundance.

3. Results and Discussion

The importance of temperature effects relative to nontemperature effects on $\Delta p\mathrm{CO}_2$ is plotted in Figure 1. As in Takahashi et al. (2002), the principal pattern is that temperature effects dominate the climatological annual mean $\Delta p\mathrm{CO}_2$ in the southern North Atlantic, while nontemperature effects (implying principally biological effects, although also advection and mixing) dominate the northern part of the basin. There is a significant degree of interannual variability in the relative importance of these effects on the annual mean $\Delta p\mathrm{CO}_2$ (Figure S5), such as in the North Atlantic Subtropical Gyre (West), which varies from a slight dominance of temperature effects (2003) to a very strong dominance (2005).

3.1. Seasonal Time Scales

To explore further the role of biological factors on $\Delta p\mathrm{CO}_2$ at seasonal scales, the results of the X-11 analysis are displayed in Figure 2. On seasonal time scales, periods of seasonally cooler water are expected to have reduced $\Delta p\mathrm{CO}_2$ in the absence of changes in DIC or alkalinity, that is, a positive correlation with SST. This is confirmed in subtropical regions; however, subpolar regions show negative correlation, implying that $\Delta p\mathrm{CO}_2$ becomes more negative in periods of seasonally warmer water, thus promoting oceanic CO_2 uptake (Figure 2a). Therefore, ocean temperature appears to be the dominant factor controlling seasonal variability in the subtropics; however, other factors (likely dominated by biological activity) appear to be more important for $\Delta p\mathrm{CO}_2$ seasonality in the subpolar region, consistent with the results of the Takahashi et al. (2002) approach (Figure 1).

The correlation of the X-11 seasonal component of $\Delta p\mathrm{CO}_2$ with PP, PIC, and NCP further supports the conclusion that $\Delta p\mathrm{CO}_2$ variability is dominated by biological activity in subpolar regions (Figures 2b–2d). Throughout the North Atlantic, and particularly in subpolar areas, seasonal increases in PP, NCP, and PIC are associated with more negative $\Delta p\mathrm{CO}_2$, suggesting increased oceanic CO_2 uptake due to biological activity. Partial correlation analysis demonstrates that this result is generally not due to the confounding effects of SST on NCP and $\Delta p\mathrm{CO}_2$ (with the exception of the North Atlantic Subtropical Gyre East province); that is, the correlation between NCP or PP and $\Delta p\mathrm{CO}_2$ is not due to a correlation between NCP or PP and SST, which itself is strongly correlated with $\Delta p\mathrm{CO}_2$. A similar partial correlation result is found for PIC, that is, that the correlation between PIC and $\Delta p\mathrm{CO}_2$ is not solely due to correlation between PIC and NCP, which in turn alters

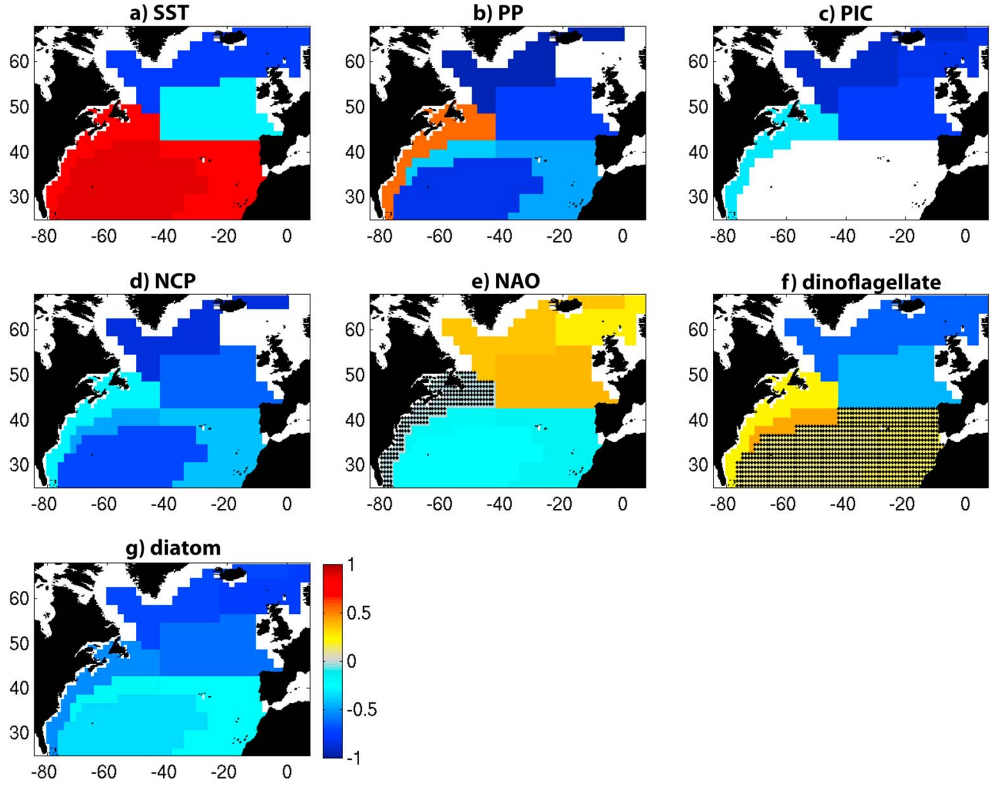

Figure 2. Correlation coefficient of the seasonal component of X-11 analysis for $\Delta p\mathrm{CO}_2$ against (a) sea surface temperature (SST), (b) primary production (PP), (c) particulate inorganic carbon (PIC), (d) net community production (NCP), (e) North Atlantic Oscillation (NAO) index, (f) dinoflagellate abundance, and (g) diatom abundance, calculated for individual provinces. Speckled areas indicate that the correlation is not statistically significant at the 95% level. For PIC, X-11 analysis was only undertaken in four provinces (see section 2).

$\Delta p\mathrm{CO}_2$. An exception is the Atlantic Arctic province, in which PIC is not significantly correlated with $\Delta p\mathrm{CO}_2$ when NCP is taken into account; that is, in this case the apparent correlation arises because PIC is correlated with NCP, which itself is correlated with $\Delta p\mathrm{CO}_2$, rather than from a direct correlation between PIC and $\Delta p\mathrm{CO}_2$. The general finding that increased PIC is associated with an increased sink after correcting for correlation with NCP is surprising, given that precipitation of 1 mol of CaCO_3 during calcification releases ~ 0.6 mol of CO_2 into the water (Frankignoulle et al., 1994). On a longer time scale, we expect the export of CaCO_3 to result in a reduction in surface $p\mathrm{CO}_2$ through ballasting (Engel et al., 2009). This effect occurs on time scales much less than a year, so it may dominate the seasonal variability but be eroded by air-sea exchange on interannual time scales (see the next section), allowing currently unknown longer-term effects to dominate the variability. In subpolar regions, biological factors appear to dominate seasonal variability in $\Delta p\mathrm{CO}_2$ in contrast to the subtropical North Atlantic, where temperature effects override biological influences at the seasonal time scale.

In addition to the role of calcifiers (represented here by PIC), we investigated the influence of other major phytoplankton groups: diatoms and dinoflagellates. The seasonal component of $\Delta p\mathrm{CO}_2$ is negatively correlated with total diatom and dinoflagellate abundance in the subpolar North Atlantic (Figures 2f and 2g), suggesting that increased abundance of both functional types is associated with increased ocean CO_2 uptake. The exception is in the northwest Atlantic, where dinoflagellate abundance is positively correlated with $\Delta p\mathrm{CO}_2$. Diatoms are traditionally thought to dominate both the subpolar North Atlantic spring bloom and the downward flux of particulate organic carbon to the deep ocean (Michaels & Silver, 1988). The negative correlation between seasonal variability in $\Delta p\mathrm{CO}_2$ and diatom abundance thus fits this canonical view. However, the negative correlation between the seasonal component of $\Delta p\mathrm{CO}_2$ and dinoflagellate abundance is of similar magnitude to that of diatoms. Dinoflagellates are not traditionally thought to contribute significantly to sinking organic carbon flux, although there is some evidence that anomalously high dinoflagellate abundance is associated with increased deep carbon flux (Henson et al., 2012). A partial correlation analysis of dinoflagellate abundance against $\Delta p\mathrm{CO_2}$ while controlling for diatom abundance confirms that dinoflagellate abundance is directly correlated with $\Delta p\mathrm{CO_2}$ (i.e., the correlation does not arise just because dinoflagellate abundance is correlated with diatom abundance, which itself is correlated with $\Delta p\mathrm{CO_2}$). Our analysis suggests therefore that it is not necessarily the relative abundance of one phytoplankton functional type or another that covaries with $\Delta p\mathrm{CO_2}$ but rather the existence (or lack) of a vigorous spring bloom (within which a progression of functional types may occur), as reflected in the negative correlation of $\Delta p\mathrm{CO_2}$ with PP, NCP, and PIC.

The NAO is positively correlated with $\Delta p\mathrm{CO_2}$ on seasonal time scales in subpolar regions and negatively in the subtropics (Figure 2e). The dominant time scale for NAO influence on ocean circulation is interannual; however, the monthly NAO index also reflects shorter time scale variability in wind patterns. Partial correlation analysis reveals that the apparent correlation between NAO and $\Delta p\mathrm{CO_2}$ in the subtropics is not significant if the effect of SST is taken into account (i.e., the correlation arises because NAO is correlated with SST, which in turn is correlated with $\Delta p\mathrm{CO_2}$). However, in the North Atlantic Drift and Atlantic Arctic provinces, NAO and $\Delta p\mathrm{CO_2}$ are significantly correlated, even accounting for SST; that is, positive NAO conditions result in increased $\Delta p\mathrm{CO_2}$ (conducive to reduced ocean uptake) in the subpolar North Atlantic. However, a positive NAO index is generally associated with stronger westerlies and therefore more rapid air-sea gas exchange, as well as cooler water temperatures at high latitudes (Visbeck et al., 2003). Both more rapid air-sea gas exchange and cooler SST would act to decrease $\Delta p\mathrm{CO_2}$ on seasonal time scales. This is in direct contrast to our results, further supporting our conclusion that temperature is not the dominant effect controlling air-sea $\mathrm{CO_2}$ flux in the subpolar region. Productivity is also reduced during positive NAO conditions (Henson et al., 2009), and mixed layer depths during winter may be deeper (Hurrell & Deser, 2009), both of which could result in increased $\Delta p\mathrm{CO_2}$. Previous work identified a potential negative correlation between coccolithophore abundance in the North Atlantic and NAO (Shutler et al., 2013), but this signal was not evident in the subpolar gyre. Collectively, these observed patterns suggest that at seasonal time scales, biological activity dominates over temperature effects in the subpolar North Atlantic. The potential role of physical processes other than temperature changes are considered in section 3.

3.2. Interannual Time Scales

A key question is whether the processes that control $\Delta p\mathrm{CO_2}$ at seasonal time scales are the same as those operating at interannual time scales. The decomposition analysis shows that the clear patterns conspicuous at seasonal scales are not necessarily evident at the interannual scale (compare Figures 2 and 3). Generally, the patterns of positive and negative correlations of $\Delta p\mathrm{CO_2}$ with the various potential controlling factors are inconsistent between the seasonal and interannual components. For example, the clear division between subpolar and subtropical regions in $\Delta p\mathrm{CO_2}$ response to SST at seasonal scales is no longer evident at interannual time scales. The exception is PP for which negative correlations with $\Delta p\mathrm{CO_2}$ exist throughout the North Atlantic at all time scales. On interannual time scales, SST is positively correlated, and NCP is negatively correlated, with $\Delta p\mathrm{CO_2}$ in subpolar regions. Therefore, SST and NCP appear to compete to alter $\Delta p\mathrm{CO_2}$. This contrasts with the findings at seasonal scales that imply that the temperature effect on $\Delta p\mathrm{CO_2}$ is secondary to biological effects. The differences in spatial patterns between Figures 2 and 3 suggest that the processes affecting $\Delta p\mathrm{CO_2}$ at time scales exceeding 1 year differ from those at the seasonal scale.

An additional example of different mechanisms working on different time scales is that of the NAO index. On seasonal time scales, NAO is positively correlated with $\Delta p\mathrm{CO_2}$ in the North Atlantic Drift Province; however, on interannual time scales, NAO is negatively correlated with $\Delta p\mathrm{CO_2}$ in the same region. How can this apparent contradiction be reconciled? The answer may lie in the different time scales on which the mechanisms affecting $\Delta p\mathrm{CO_2}$ operate. Seasonally, positive NAO conditions are associated with reduced PP in the subpolar North Atlantic due to stronger winds and deeper mixing (Henson et al., 2009). Despite lower SST in positive NAO periods, the overall effect is to reduce PP, which, on a seasonal time scale, acts to reduce ocean uptake. However, at the interannual scale, positive NAO periods are associated with increased ocean carbon uptake (decreased $\Delta p\mathrm{CO_2}$) in the Northeast Atlantic due to intensified advection of waters low in DIC in the North Atlantic current from the subtropics (Thomas et al., 2008). This disparity in the association between NAO and $\Delta p\mathrm{CO_2}$ over different time scales is clearly shown in our analysis. The decomposition method used

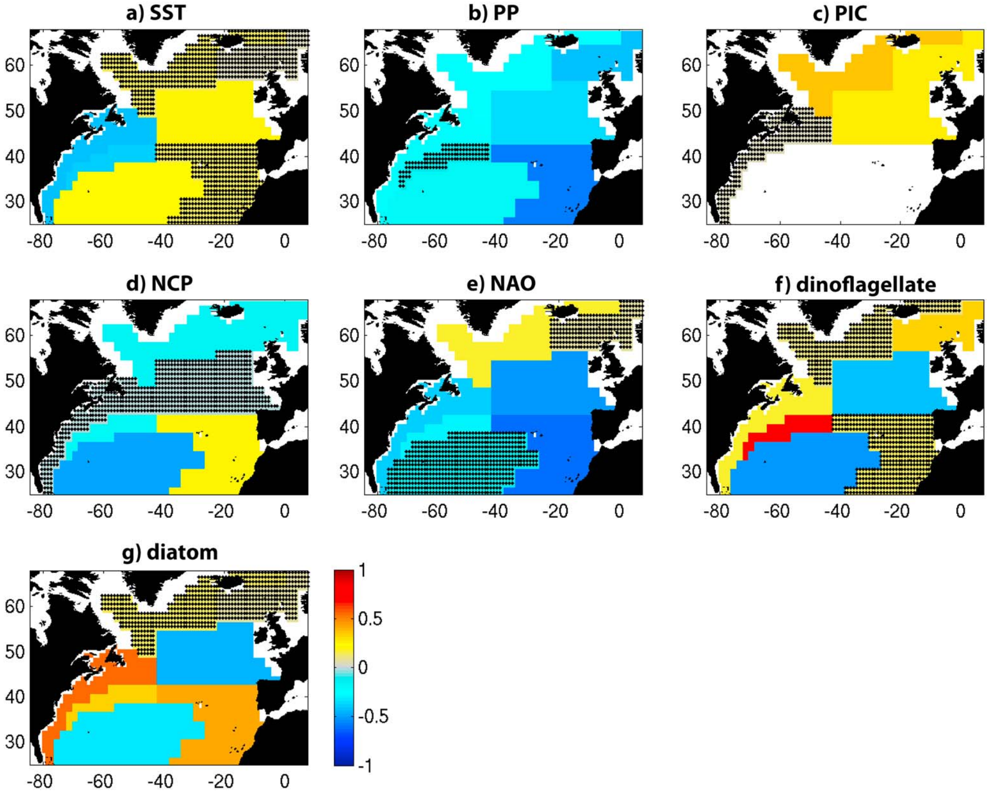

Figure 3. As in Figure 2 but showing the correlation coefficient of the interannual component of the X-11 analysis for $\Delta p\text{CO}_2$ against potential controls.

here therefore allows novel insights into how the factors controlling $\Delta p\text{CO}_2$ may depend on the time scale under consideration.

The equilibration time scale of CO_2 between the surface ocean and the atmosphere is ~ 6 months to 1 year (Jones et al., 2014); at longer than seasonal time scales, air-sea exchange erodes the $\Delta p\text{CO}_2$ signal established by seasonal biological or temperature variability. Halloran et al. (2015) identify four mechanisms hypothesized to control variability in ocean CO_2 uptake in the North Atlantic on decadal time scales: biological activity, temperature, vertical mixing, and horizontal advection. For example, increased intensity of deep convection, prevalent in the Labrador Sea (Pickart et al., 2003), increases surface DIC but also introduces additional nutrients, promoting biological carbon export (Ullman et al., 2009). Additionally, changes in circulation can alter horizontal advection, affecting transport of DIC or total alkalinity (Corbiere et al., 2007). In our analysis, use of large-scale provinces blurs somewhat any potential influence of advection-driven changes in $\Delta p\text{CO}_2$. However, we note that, on time scales exceeding 1 year, changes in vertical mixing or horizontal transport, in addition to temperature and biological effects, are likely to be significant (Gruber, 2009).

4. Conclusion

The analysis presented here uncovers novel insights into potential controls on North Atlantic $\Delta p\text{CO}_2$ by separating seasonal and interannual time scales. On seasonal time scales, we find the expected pattern of temperature dominance on $\Delta p\text{CO}_2$ in the subtropics and PP dominance at high latitudes. However, at time scales exceeding 1 year, temperature effects also become important at high latitudes, and the role of biological processes becomes less clear. The decomposition used here clarifies that the NAO influences $\Delta p\text{CO}_2$ in subpolar regions on seasonal time scales (potentially via altering NCP), but we expect that advective effects are likely to be more important on interannual scales. We also conclude that the presence of a robust bloom (regardless of its composition) is likely important in controlling $\Delta p\text{CO}_2$.

Our analysis shows that understanding the mechanisms underlying seasonal variability in $\Delta p\mathrm{CO}_2$ does not directly inform on how the North Atlantic CO_2 sink responds to interannual forcing. Mechanistic understanding of the North Atlantic CO_2 sink should not therefore be based solely on seasonal drivers but should also consider interannual variability. At decadal time scales the processes affecting $\Delta p\mathrm{CO}_2$ may be different again and principally associated with ocean circulation and ventilation, as reflected in large-scale climate modes such as the Atlantic Multidecadal Oscillation (McKinley et al., 2017). In the North Pacific, SST and advection dominate variability in $\Delta p\mathrm{CO}_2$ at seasonal scales (Takahashi et al., 2009), although biology also plays a role (Ayers & Lozier, 2012). However, on decadal scales the Pacific Decadal Oscillation is the dominant driver via its effects on SST and mixed layer depth (Yasunaka et al., 2014). In the Southern Ocean, the Southern Annular Mode is highly correlated with $\Delta p\mathrm{CO}_2$ variability on the interannual scale due to its influence on westerly winds and upwelling of DIC-rich waters (Lovenduski et al., 2007); however, at the decadal scale Southern Annular Mode is no longer the principal driver (Fay & McKinley, 2013). Except for the equatorial Pacific region, climate oscillations explain only a small fraction of $\Delta p\mathrm{CO}_2$ variability (Breeden & McKinley, 2016), illustrating the importance of other controlling factors. A full understanding of how the various forcing factors may combine to drive $\Delta p\mathrm{CO}_2$ in all oceans will only be possible with long-term, consistent time series of observations. We note also that the choice of PP or NCP data set may influence the patterns of correlation with $\Delta p\mathrm{CO}_2$ described here (supporting information). There remains uncertainty therefore about the mechanisms underpinning seasonal and interannual variability in $\Delta p\mathrm{CO}_2$, which underscores the need for continued long-term multiyear observations of the global marine carbon cycle. The current lack of understanding limits our ability to model the global oceanic sink and thus reliably predict its trajectory under ongoing increases in anthropogenic CO_2.

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