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Controls on open-ocean North Atlantic $\Delta p$CO$_2$ at seasonal and interannual timescales are different

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Key points:
- Observational evidence that the mechanisms underlying seasonal variability in $\Delta p$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 $\Delta p$CO$_2$ in subpolar waters
- Long-term observations of ocean CO$_2$ are required to distinguish seasonal and interannual controls on $\Delta p$CO$_2$

Index terms:
4800 Oceanography: Biological and Chemical
4805 Biogeochemical cycles, processes, and modelling
4806 Carbon cycling
4504 Air/sea interactions
Abstract

The North Atlantic is a substantial sink for anthropogenic CO₂. 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 non-biological) that affect the sea-air CO₂ difference (ΔpCO₂) on seasonal and interannual timescales. We demonstrate that, on seasonal timescales, the subpolar North Atlantic ΔpCO₂ signal is predominantly correlated with biological processes, whereas seawater temperature dominates in the subtropics. However, the same factors do not necessarily control ΔpCO₂ on interannual timescales. Our results imply that the mechanisms driving seasonal variability in ΔpCO₂ cannot necessarily be extrapolated to predict how ΔpCO₂, and thus the North Atlantic CO₂ sink, may respond to increases in anthropogenic CO₂ over longer timescales.

Plain language summary

As atmospheric carbon dioxide (CO₂) concentrations rise due to anthropogenic emissions, the ocean is taking up more CO₂; a process known as the oceanic CO₂ sink. The North Atlantic is a major anthropogenic CO₂ 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 CO₂ sink. Often the factors that affect oceanic uptake of CO₂ 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 CO₂ uptake to seasonal variability out to longer time scales.
1. Introduction

On multi-decadal timescales the ocean is a key route for removal of anthropogenic CO₂ from the atmosphere, taking up approximately one third of emissions since pre-industrial times [Khatiwala et al., 2013]. The high latitude North Atlantic has one of the highest uptake rates of atmospheric CO₂ per square metre [Mikaloff-Fletcher et al., 2006], accounting for 23% of oceanic anthropogenic CO₂ storage, whilst only constituting 15% of the global ocean surface area [Sabine et al., 2004]. However, recent studies suggest that the North Atlantic CO₂ 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 CO₂ 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 CO₂ concentration difference across the boundary layer determines the net direction of CO₂ transfer [Woolf et al., 2016], i.e. the difference between the partial pressure of CO₂ (pCO₂) in seawater and the overlying atmosphere (ΔpCO₂). 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 CO₂ transfer. Where ΔpCO₂ is positive (seawater pCO₂ > atmospheric pCO₂), the water is oversaturated, implying a net flux from sea to air, i.e. a potential CO₂ ‘source’. The opposite case, where ΔpCO₂ is negative and the ocean is undersaturated, implies a CO₂ ‘sink’. Atmospheric pCO₂ is homogeneous relative to seawater, so seawater pCO₂ is typically the dominant control on ΔpCO₂ direction. Thus, biogeochemical and hydrographic processes can modify ΔpCO₂ if they alter the seawater pCO₂. Cooler water has a greater capacity to store dissolved inorganic carbon (DIC) than warm water, as CO₂ solubility is inversely proportional to water temperature. Cooler water reduces seawater pCO₂, helping to drive negative ΔpCO₂, while warming has the opposite effect. Net community production (NCP, primary production minus respiration) takes up DIC from the seawater through photosynthesis, decreasing seawater pCO₂ and contributing to negative ΔpCO₂. Calcification consumes DIC, but is a CO₂ source due to the accompanying net release of CO₂ into the water [Frankignoule et al., 1994], which may have a significant localised impact in the North Atlantic [Shutler et al., 2013]. The net effect of the combination of physical and biological drivers results in an overall CO₂ 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: phyttoplankton community structure [Hilligsøe et al., 2011], with functional types such as diatoms, which are thought to export organic carbon most efficiently, expected to have a dominant effect [Michaels and Silver, 1988]. However, small phytoplankton have also been found to influence CO$_2$ uptake and export [Palevsky et al., 2013; Richardson and Jackson, 2007] and in the North Atlantic dinoflagellate abundance was found to strongly correlate with organic carbon flux at 2000 m [Henson et al., 2012]. Whilst calcifying phytoplankton (e.g. coccolithophores) can also modify seawater $p$CO$_2$ during formation, they may also contribute to efficient organic carbon transfer to depth [Klaas and Archer, 2002].

The potential controls on the North Atlantic CO$_2$ sink at different timescales are not well understood. For example, in a model study Bennington et al. [2009] found that biological activity dominated the seasonal cycle of seawater $p$CO$_2$, but not its interannual variability. On these longer timescales, 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 datasets we examine the hypothesis, suggested by a previous model study [Bennington et al., 2009], that the dominant influences on $\Delta p$CO$_2$ in the North Atlantic are different at seasonal and interannual timescales. One approach is to separate the effects using a climatological mass balance technique [Ayers and Lozier, 2012], however to specifically test the importance of potential mechanisms at different timescales, 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 $\Delta p$CO$_2$ are potentially valid on both seasonal and longer timescales e.g. that biological effects dominate over temperature effects at high latitudes.
2. Methods

Monthly gridded fugacity of seawater CO₂ ($f$CO₂) for the North Atlantic was downloaded from the SOCAT v3 database (Bakker et al., 2016; www.socat.info) and re-analysed to a common sea surface temperature (SST) dataset [Reynolds et al., 2007] at 0.5 m depth, following the method of Goddijn-Murphy et al. [2015]. Ocean $p$CO₂ was calculated from $f$CO₂ using the equations and constants provided in the seacarb R package v3 [Lavigne et al., 2011]. The data were then gridded to a 1x1° grid following the SOCAT method [Sabine et al., 2013]. To calculate the Δ$p$CO₂, atmospheric molar CO₂ concentration was obtained from the NOAA Marine Boundary Layer reference dataset (https://www.esrl.noaa.gov/gmd/ccgg/mbl/index.html). These were converted to $p$CO₂(air) using the formulation of Weiss [1974], as implemented in Shutler et al. [2016]. Auxiliary datasets for sea surface salinity and sea level pressure were taken from the World Ocean Atlas 2013 climatology [Zweng et al., 2013] and the NCEP/NCAR Reanalysis dataset [Kalnay et al., 1996; http://www.esrl.noaa.gov/psd/], respectively. MODIS-Aqua chlorophyll concentration, photosynthetically available radiation (PAR), particulate inorganic carbon (PIC) and night-time 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 PAR data to estimate primary production (PP) using the Vertically Generalised Productivity Model [Behrenfeld and Falkowski, 1997]. PIC data were corrected for sensor saturation effects by filling erroneously missing data [Land et al., 2017]. Monthly satellite-derived NCP estimates were taken from Tilstone et al. [2015]. All satellite-derived data were re-gridded onto a 1x1° grid to match the resolution of the Δ$p$CO₂ 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 (CPR) survey (Richardson et al., 2006; dataset doi: 10.7487/2016.194.1.988). MODIS data were available for the time period July 2002-December 2014, which is used for all analyses with the exception of NCP data which ends in 2010. We repeated the analysis for 3 alternative PP algorithms [Westberry et al., 2008; Marra et al., 2003; Carr, 2001] and 2 alternative NCP algorithms [Li and Cassar, 2016; Siegel et al., 2014] to investigate the sensitivity of our results to the choice of satellite PP and NCP algorithms (supplementary material). For PP, the 3 chlorophyll-based algorithms (VGPM, Marra and Carr) all agree on the pattern of correlation with Δ$p$CO₂ at both seasonal and interannual timescales. Although the CbPM model [Westberry et al.,
2008] displays negative correlation between PP and \( \Delta pCO_2 \) in the subpolar region at seasonal timescales (consistent with other algorithms tested), in the subtropics and at interannual timescales CbPM-PP is positively correlated with \( \Delta pCO_2 \) (differing from other algorithms tested). For NCP at the seasonal timescale, 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 mid-latitude provinces. At the interannual timescale, both the Li and Cassar [2016] and Siegel et al. [2014] algorithms have several regions where NCP and \( \Delta pCO_2 \) 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, nevertheless these are the only PP and NCP estimates available at the basin-scale and multi-year timescale which are essential for our analysis.

To overcome limitations in the spatial coverage of the SOCAT database observations from 2002-2014, \( \Delta pCO_2 \) was averaged within Longhurst provinces [Longhurst, 1998], as were all satellite-derived and CPR data. Provinces in which \( >65\% \) of the \( \Delta pCO_2 \) time series had missing data were excluded, as were those encompassing shelf regions. In the remaining regions, any missing province-mean monthly \( \Delta pCO_2 \) 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 \( pCO_2 \) change into temperature driven and non-temperature driven effects. The non-temperature driven term is characterised as the “net biology” effect [Takahashi et al., 2002], which includes net PP, net alkalinity change due to nutrient utilisation, 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 sub-surface waters. Although the non-temperature effects are dominated by biological activity, this approach is not able to distinguish the type of biological effect, e.g. due to community metabolism or calcification. Therefore, to identify potential dominant biological effects we also analyse 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 \( \Delta pCO_2 \), SST, PP, NCP, PIC, and diatom and dinoflagellate abundance spatially averaged over Longhurst provinces.

An example of the decomposition generated by the X-11 method is given in Figure S4. The raw time series of \( \Delta pCO_2 \), 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 non-parametric Spearman correlation coefficient between the different components of \( \Delta pCO_2 \) 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 non-parametric partial correlation coefficients to assess association between \( \Delta pCO_2 \) components and potential drivers while controlling for the effect of other variables (e.g. Brown and Hendrix, 2014). For example, we wish to investigate whether the seasonal component of \( \Delta pCO_2 \) is correlated with PIC, however PIC is also potentially correlated with NCP which is itself correlated with \( \Delta pCO_2 \). Partial correlation analysis allows us to determine whether PIC is statistically significantly correlated with \( \Delta pCO_2 \) whilst controlling for the effect of NCP. We also test the correlation between \( \Delta pCO_2 \) and NCP, and between \( \Delta pCO_2 \) and NAO while controlling for SST, and the correlation between \( \Delta pCO_2 \) and dinoflagellate abundance while controlling for diatom abundance.

3. Results and Discussion
The importance of temperature effects relative to non-temperature effects on $\Delta p\text{CO}_2$ is plotted in Figure 1. As in Takahashi et al. [2002], the principal pattern is that temperature effects dominate the climatological annual mean $\Delta p\text{CO}_2$ in the southern North Atlantic, while non-temperature 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 $\Delta p\text{CO}_2$ (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 timescales

To explore further the role of biological factors on $\Delta p\text{CO}_2$ at seasonal scales, the results of the X-11 analysis are displayed in Figure 2. On seasonal timescales, periods of seasonally cooler water are expected to have reduced $\Delta p\text{CO}_2$ in the absence of changes in DIC or alkalinity, i.e. a positive correlation with SST. This is confirmed in subtropical regions, however subpolar regions show negative correlation, implying that $\Delta p\text{CO}_2$ 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 $\Delta p\text{CO}_2$ 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 $\Delta p\text{CO}_2$ with PP, PIC and NCP further supports the conclusion that $\Delta p\text{CO}_2$ variability is dominated by biological activity in subpolar regions (Figure 2b,c,d). Throughout the North Atlantic, and particularly in subpolar areas, seasonal increases in PP, NCP and PIC are associated with more negative $\Delta p\text{CO}_2$, 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 $\Delta p\text{CO}_2$ (with the exception of the North Atlantic Subtropical Gyre East province), i.e. the correlation between NCP or PP and $\Delta p\text{CO}_2$ is not due to a correlation between NCP or PP and SST, which itself is strongly correlated with $\Delta p\text{CO}_2$. A similar partial correlation result is found for PIC, i.e. that the correlation between PIC and $\Delta p\text{CO}_2$ is not solely due to correlation between PIC and NCP, which in turn alters $\Delta p\text{CO}_2$. An exception is the Atlantic Arctic province, in which PIC is not significantly correlated with $\Delta p\text{CO}_2$ when NCP is taken into account, i.e. in this case the apparent correlation arises
because PIC is correlated with NCP, which itself is correlated with $\Delta p$CO$_2$, rather than from a direct correlation between PIC and $\Delta p$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 one mole of CaCO$_3$ during calcification releases ~ 0.6 mole of CO$_2$ into the water [Frankignoulle et al., 1994]. On a longer timescale, we expect the export of CaCO$_3$ to result in a reduction in surface $p$CO$_2$ through ballasting [Engel et al., 2009]. This effect occurs on timescales much less than a year, so it may dominate the seasonal variability but be eroded by air-sea exchange on interannual timescales (see next section), allowing currently unknown longer-term effects to dominate the variability. In subpolar regions, biological factors appear to dominate seasonal variability in $\Delta p$CO$_2$ in contrast to the subtropical North Atlantic, where temperature effects override biological influences at the seasonal timescale.

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 $\Delta p$CO$_2$ is negatively correlated with total diatom and dinoflagellate abundance in the subpolar North Atlantic (Figure 2f,g), 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 $\Delta p$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 and Silver, 1998]. The negative correlation between seasonal variability in $\Delta p$CO$_2$ and diatom abundance thus fits this canonical view. However, the negative correlation between the seasonal component of $\Delta p$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 $\Delta p$CO$_2$ whilst controlling for diatom abundance confirms that dinoflagellate abundance is directly correlated with $\Delta p$CO$_2$ (i.e. the correlation does not arise just because dinoflagellate abundance is correlated with diatom abundance, which itself is correlated with $\Delta p$CO$_2$). Our analysis suggests therefore that it is not necessarily the relative abundance of one phytoplankton functional type or another that covaries with $\Delta p$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 $\Delta pCO_2$ with PP, NCP and PIC.

The NAO is positively correlated with $\Delta pCO_2$ on seasonal timescales in subpolar regions, and negatively in the subtropics (Figure 2e). The dominant timescale for NAO influence on ocean circulation is interannual, however the monthly NAO index also reflects shorter timescale variability in wind patterns. Partial correlation analysis reveals that the apparent correlation between NAO and $\Delta pCO_2$ 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 $\Delta pCO_2$). However, in the North Atlantic Drift and Atlantic Arctic provinces, NAO and $\Delta pCO_2$ are significantly correlated, even accounting for SST, i.e. positive NAO conditions result in increased $\Delta pCO_2$ (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 $\Delta pCO_2$ on seasonal timescales. This is in direct contrast to our results, further supporting our conclusion that temperature is not the dominant effect controlling air-sea 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 and Deser, 2009], both of which could result in increased $\Delta pCO_2$. 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 timescales, biological activity dominates over temperature effects in the subpolar North Atlantic. The potential role of physical processes other than temperature changes are considered in the discussion section.

### 3.2 Interannual timescales

A key question is whether the processes that control $\Delta pCO_2$ at seasonal timescales are the same as those operating at interannual timescales. 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 correlation of $\Delta pCO_2$ 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 $\Delta pCO_2$ response to SST at seasonal scales is no longer evident at interannual timescales. The exception is PP for which negative correlations with $\Delta pCO_2$ exist throughout the North Atlantic at all timescales. On interannual timescales, SST is positively correlated, and NCP is negatively correlated, with $\Delta pCO_2$ in subpolar regions. Therefore, SST and NCP appear to compete to alter $\Delta pCO_2$. This contrasts with the findings at seasonal scales that imply the temperature effect on $\Delta pCO_2$ is secondary to biological effects. The differences in spatial patterns between Figures 2 and 3 suggest that the processes affecting $\Delta pCO_2$ at timescales exceeding one year differ from those at the seasonal scale.

An additional example of different mechanisms working on different timescales is that of the NAO index. On seasonal timescales, NAO is positively correlated with $\Delta pCO_2$ in the North Atlantic Drift Province; however on interannual timescales, NAO is negatively correlated with $\Delta pCO_2$ in the same region. How can this apparent contradiction be reconciled? The answer may lie in the different timescales on which the mechanisms affecting $\Delta pCO_2$ 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 timescale, acts to reduce ocean uptake. However, at the interannual scale, positive NAO periods are associated with increased ocean carbon uptake (decreased $\Delta pCO_2$) 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 $\Delta pCO_2$ over different time scales is clearly shown in our analysis. The decomposition method used here therefore allows novel insights into how the factors controlling $\Delta pCO_2$ may depend on the timescale under consideration.

The equilibration timescale of CO$_2$ between the surface ocean and the atmosphere is ~6 months to 1 year [Jones et al., 2014]; at longer than seasonal timescales, air-sea exchange erodes the $\Delta pCO_2$ signal established by seasonal biological or temperature variability. Halloran et al. [2015] identify four mechanisms hypothesised to control variability in ocean CO$_2$ uptake in the North Atlantic on decadal timescales: 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 $\Delta pCO_2$. However, we note that, on timescales exceeding one 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 $\Delta pCO_2$ by separating seasonal and interannual timescales. On seasonal timescales, we find the expected pattern of temperature dominance on $\Delta pCO_2$ in the subtropics, and PP dominance at high latitudes. However, at timescales exceeding one 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 $\Delta pCO_2$ in subpolar regions on seasonal timescales (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 $\Delta pCO_2$.

Our analysis shows that understanding the mechanisms underlying seasonal variability in $\Delta pCO_2$ 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 timescales the processes affecting $\Delta pCO_2$ 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 $\Delta pCO_2$ at seasonal scales [Takahashi et al., 2009], although biology also plays a role [Ayers and 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 (SAM) is highly correlated with $\Delta pCO_2$ 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 SAM is no longer the principal driver [Fay and McKinley, 2013]. Except for the equatorial Pacific region, climate oscillations explain only a small fraction of $\Delta pCO_2$ variability [Breeden and McKinley, 2016], illustrating the importance of other controlling
factors. A full understanding of how the various forcing factors may combine to drive \(\Delta p\text{CO}_2\) 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 dataset may influence the patterns of correlation with \(\Delta p\text{CO}_2\) described here (supplementary material). There remains uncertainty therefore about the mechanisms underpinning seasonal and interannual variability in \(\Delta p\text{CO}_2\), which underscores the need for continued long-term multi-year 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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**Figure Captions**

**Figure 1**: Relative importance of temperature and net biological effects on climatological ΔpCO₂ based on the decomposition of Takahashi et al. [2002]. Provinces are defined and named as per Longhurst [1998].

**Figure 2**: Correlation coefficient of the seasonal component of X-11 analysis for ΔpCO₂ against a) SST, b) primary production, c) particulate inorganic carbon, d) net community production, e) North Atlantic Oscillation 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 4 provinces (see Methods).

**Figure 3**: As for Figure 2, but showing the correlation coefficient of the interannual component of the X-11 analysis for ΔpCO₂ against potential controls.