



RESEARCH LETTER

10.1029/2018GL078797

Key Points:

- Observational evidence shows 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 ΔpCO₂ in subpolar waters
- Long-term observations of ocean CO_2 are required to distinguish seasonal and interannual controls on ΔpCO_2

Supporting Information: • Supporting Information S1

Correspondence to:

S. A. Henson, s.henson@noc.ac.uk

Citation:

Henson, S. A., Humphreys, M. P., Land, P. E., Shutler, J. D., Goddijn-Murphy, L., & Warren, M. (2018). Controls on open-ocean North Atlantic ΔpCO_2 at seasonal and interannual time scales are different. *Geophysical Research Letters*, 45. https://doi.org/10.1029/ 2018GL078797

Received 16 MAY 2018 Accepted 30 JUL 2018 Accepted article online 6 AUG 2018

©2018. The Authors.

This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

Controls on Open-Ocean North Atlantic ΔpCO_2 at Seasonal and Interannual Time Scales Are Different

Stephanie A. Henson¹, Matthew P. Humphreys^{2,3}, Peter E. Land⁴, Jamie D. Shutler⁵, Lonneke Goddijn-Murphy⁶, and Mark Warren⁴

¹National Oceanography Centre, Southampton, UK, ²Ocean and Earth Science, University of Southampton, Southampton, UK, ³Now at School of Environmental Sciences, University of East Anglia, Norwich, UK, ⁴Plymouth Marine Laboratory, Plymouth, UK, ⁵College of Life and Environmental Sciences, University of Exeter, Penryn, UK, ⁶Environmental Research Institute, University of the Highlands and Islands, Thurso, UK

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 nonbiological) that affect the sea-air CO₂ difference (ΔpCO_2) on seasonal and interannual time scales. We demonstrate that on seasonal time scales, the subpolar North Atlantic ΔpCO_2 signal is predominantly correlated with biological processes, whereas seawater temperature dominates in the subtropics. However, the same factors do not necessarily control ΔpCO_2 on interannual time scales. Our results imply that the mechanisms driving seasonal variability in ΔpCO_2 cannot necessarily be extrapolated to predict how ΔpCO_2 , and thus the North Atlantic CO₂ sink, may respond to increases in anthropogenic CO₂ over longer time scales.

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 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 CO₂ concentration difference across the boundary layer determines the net direction of CO₂ transfer (Woolf et al., 2016), that is, the difference between the partial pressure of CO₂ (pCO₂) in seawater and the overlying atmosphere (Δp CO₂). 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 Δp CO₂ is positive (seawater pCO₂ > atmospheric pCO₂), the water is oversaturated, implying a net flux from sea to air, that is, a potential CO₂ "source." The opposite case, where Δp CO₂ 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 Δp CO₂ direction. Thus, biogeochemical and hydrographic processes can modify Δp CO₂ if they alter the seawater pCO₂. Cooler



<mark>-</mark>

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 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 CO₂ source due to the accompanying net release of CO₂ 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 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: 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 CO₂ (fCO₂) for the North Atlantic was downloaded from the Surface Ocean CO₂ 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 CO₂ 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 $1 \times 1^{\circ}$ grid to match the resolution of the ΔpCO_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 ΔpCO_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 $\Delta p CO_2$ 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 ΔpCO_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 ΔpCO_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, ΔpCO_2 was averaged within Longhurst provinces (Longhurst, 1998), as were all satellite-derived and Continuous Plankton Recorder data. Provinces in which >65% of the ΔpCO_2 time series had missing data were excluded, as were those encompassing shelf regions. In the remaining regions, any missing provincemean monthly ΔpCO_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 temperaturedriven 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 ΔpCO_2 , SST, PP, NCP, PIC, and diatom and dinoflagellate abundance spatially averaged over Longhurst provinces.



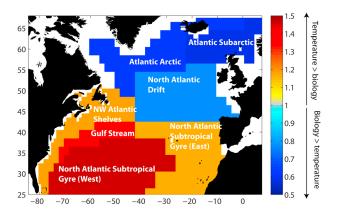


Figure 1. Relative importance of temperature and net biological effects on climatological ΔpCO_2 based on the decomposition of Takahashi et al. (2002). Provinces are defined and named as per Longhurst (1998).

An example of the decomposition generated by the X-11 method is given in Figure S4. The raw time series of ΔpCO_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 ΔpCO_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 ΔpCO_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 ΔpCO_2 is correlated with PIC; however, PIC is also potentially correlated with NCP, which is itself correlated with ΔpCO_2 . Partial correlation analysis allows us to determine whether PIC is statistically significantly correlated with ΔpCO_2 while controlling for the effect of NCP. We also test the correlation between ΔpCO_2 and NCP, and between ΔpCO_2 and NAO while controlling for SST, and the correlation between ΔpCO_2 and dinoflagellate abundance while controlling for diatom abundance.

3. Results and Discussion

The importance of temperature effects relative to nontemperature effects on ΔpCO_2 is plotted in Figure 1. As in Takahashi et al. (2002), the principal pattern is that temperature effects dominate the climatological annual mean ΔpCO_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 ΔpCO_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 ΔpCO_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 ΔpCO_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 ΔpCO_2 becomes more negative in periods of seasonally warmer water, thus promoting oceanic CO₂ 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 ΔpCO_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 ΔpCO_2 with PP, PIC, and NCP further supports the conclusion that ΔpCO_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 ΔpCO_2 , suggesting increased oceanic CO₂ 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 ΔpCO_2 (with the exception of the North Atlantic Subtropical Gyre East province); that is, the correlation between NCP or PP and ΔpCO_2 is not due to a correlation between NCP or PP and SST, which itself is strongly correlated with ΔpCO_2 is not solely due to correlation between PIC and NCP, which in turn alters

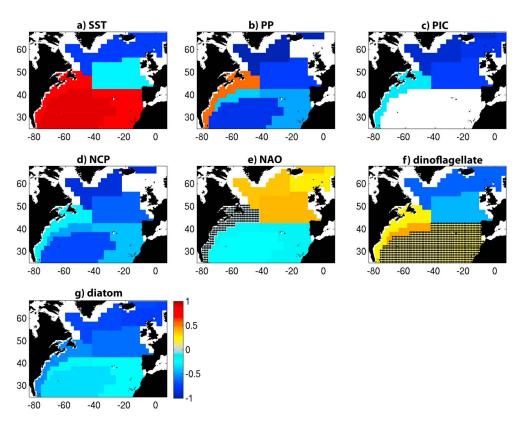


Figure 2. Correlation coefficient of the seasonal component of X-11 analysis for ΔpCO_2 against (a) sea surface temperature (SST), (b) primary production (PP), (c) particulate inorganic carbon (PIC), (d) net community production (NCP), (e) North Atlantic Oscillation (NAO) index, (f) dinoflagellate abundance, 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).

 ΔpCO_2 . An exception is the Atlantic Arctic province, in which PIC is not significantly correlated with ΔpCO_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 ΔpCO_2 , rather than from a direct correlation between PIC and ΔpCO_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 CaCO₃ during calcification releases ~ 0.6 mol of CO₂ into the water (Frankignoulle et al., 1994). On a longer time scale, we expect the export of CaCO₃ to result in a reduction in surface pCO_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 longerterm effects to dominate the variability. In subpolar regions, biological factors appear to dominate seasonal variability in ΔpCO_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 ΔpCO_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 ΔpCO_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 ΔpCO_2 and diatom abundance thus fits this canonical view. However, the negative correlation between the seasonal component of ΔpCO_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 ΔpCO_2 while controlling for diatom abundance confirms that dinoflagellate abundance is directly correlated with ΔpCO_2 (i.e., the correlation does not arise just because dinoflagellate abundance is correlated with diatom abundance, which itself is correlated with ΔpCO_2). Our analysis suggests therefore that it is not necessarily the relative abundance of one phytoplankton functional type or another that covaries with ΔpCO_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 ΔpCO_2 with PP, NCP, and PIC.

The NAO is positively correlated with $\Delta p CO_2$ 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 $\Delta p CO_2$ 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 ΔpCO_2). However, in the North Atlantic Drift and Atlantic Arctic provinces, NAO and ΔpCO_2 are significantly correlated, even accounting for SST; that is, positive NAO conditions result in increased $\Delta p CO_2$ (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 $\Delta p CO_2$ on seasonal time scales. This is in direct contrast to our results, further supporting our conclusion that temperature is not the dominant effect controlling airsea CO₂ 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 ΔpCO_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 ΔpCO_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 ΔpCO_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 ΔpCO_2 response to SST at seasonal scales is no longer evident at interannual time scales. The exception is PP for which negative correlations with ΔpCO_2 exist throughout the North Atlantic at all time scales. On interannual time scales, SST is positively correlated, and NCP is negatively correlated, with ΔpCO_2 in subpolar regions. Therefore, SST and NCP appear to compete to alter ΔpCO_2 . This contrasts with the findings at seasonal scales that imply that the temperature effect on ΔpCO_2 is secondary to biological effects. The differences in spatial patterns between Figures 2 and 3 suggest that the processes affecting ΔpCO_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 ΔpCO_2 in the North Atlantic Drift Province; however, on interannual time scales, NAO is negatively correlated with ΔpCO_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 ΔpCO_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 ΔpCO_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 ΔpCO_2 over different time scales is clearly shown in our analysis. The decomposition method used

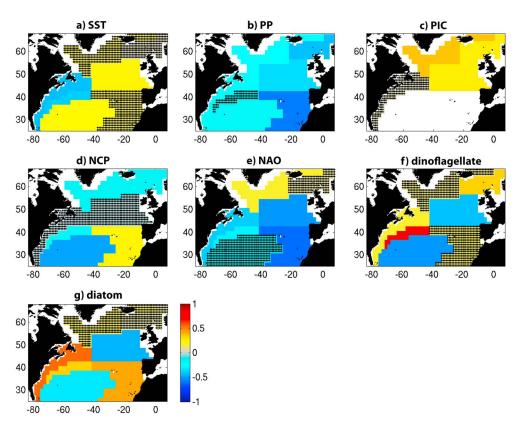


Figure 3. As in Figure 2 but showing the correlation coefficient of the interannual component of the X-11 analysis for ΔpCO_2 against potential controls.

here therefore allows novel insights into how the factors controlling ΔpCO_2 may depend on the time scale under consideration.

The equilibration time scale of CO₂ 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 Δp CO₂ signal established by seasonal biological or temperature variability. Halloran et al. (2015) identify four mechanisms hypothesized to control variability in ocean CO₂ 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 Δp CO₂. 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 ΔpCO_2 by separating seasonal and interannual time scales. On seasonal time scales, we find the expected pattern of temperature dominance on ΔpCO_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 ΔpCO_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 ΔpCO_2 .



Our analysis shows that understanding the mechanisms underlying seasonal variability in $\Delta p CO_2$ does not directly inform on how the North Atlantic CO₂ sink responds to interannual forcing. Mechanistic understanding of the North Atlantic CO₂ sink should not therefore be based solely on seasonal drivers but should also consider interannual variability. At decadal time scales the processes affecting $\Delta p CO_2$ 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 ΔpCO_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 ΔpCO_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 $\Delta p CO_2$ variability (Breeden & McKinley, 2016), illustrating the importance of other controlling factors. A full understanding of how the various forcing factors may combine to drive $\Delta p CO_2$ 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 ΔpCO_2 described here (supporting information). There remains uncertainty therefore about the mechanisms underpinning seasonal and interannual variability in ΔpCO_{2r} , 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 CO₂.

Acknowledgments

This work forms a contribution to the National Environmental Research Council program on greenhouse gases (NE/K00249X/1, NE/K002546/1, and NE/K002511/1). SOCAT is an international effort, endorsed by the International Ocean Carbon Coordination Project, the Surface Ocean Lower Atmosphere Study, and the Integrated Marine Biosphere Research programs, to deliver a uniformly qualitycontrolled surface ocean CO₂ database. The many researchers and funding agencies responsible for the collection of data and quality control are thanked for their contributions to SOCAT. The reanalyzed and gridded SOCAT data were provided by the European Space Agency project OceanFlux Greenhouse Gases Evolution (http://www.oceanfluxghg.org).

References

- Ayers, J. M., & Lozier, M. S. (2012). Unraveling dynamical controls on the North Pacific carbon sink. *Journal of Geophysical Research*, 117, C01017. https://doi.org/10.1029/2011JC007368
- Bakker, D. C. E., Pfeil, B., Landa, C. S., Metzl, N., O'Brien, K. M., Olsen, A., et al. (2016). A multi-decade record of high-quality fCO(2) data in version 3 of the Surface Ocean CO₂ Atlas (SOCAT). *Earth System Science Data*, 8(2), 383–413. https://doi.org/10.5194/essd-8-383-2016
- Behrenfeld, M. J., & Falkowski, P. G. (1997). Photosynthetic rates derived from satellite-based chlorophyll concentration. Limnology and Oceanography, 42(1), 1–20. https://doi.org/10.4319/lo.1997.42.1.0001
- Bennington, V., McKinley, G. A., Dutkiewicz, S., & Ulman, D. (2009). What does chlorophyll variability tell us about export and air-sea CO₂ flux variability in the North Atlantic? *Global Biogeochemical Cycles*, 23, GB3002. https://doi.org/10.1029/2008GB003241
- Breeden, M. L., & McKinley, G. A. (2016). Climate impacts on multidecadal pCO₂ variability in the North Atlantic: 1948-2009. *Biogeosciences*, 13(11), 3387–3396. https://doi.org/10.5194/bg-13-3387-2016
- Brown, B. L., & Hendrix, S. B. (2014). Partial correlation coefficients. In Wiley StatsRef: Statistics Reference Online. https://doi.org/10.1002/ 9781118445112.stat06488
- Carr, M.-E. (2001). Estimation of potential productivity in Eastern Boundary Currents using remote sensing. Deep Sea Research Part II: Topical Studies in Oceanography, 49(1–3), 59–80. https://doi.org/10.1016/S0967-0645(01)00094-7
- Corbiere, A., Metzl, N., Reverdin, G., Brunet, C., & Takahashi, A. (2007). Interannual and decadal variability of the oceanic carbon sink in the North Atlantic subpolar gyre. *Tellus Series B: Chemical and Physical Meteorology*, 59(2), 168–178. https://doi.org/10.1111/j.1600-0889.2006.00232.x
- Engel, A., Szlosek, J., Abramson, L., Liu, Z., & Lee, C. (2009). Investigating the effect of ballasting by CaCO₃ in Emiliania huxleyi: I. Formation, settling velocities and physical properties of aggregates. *Deep Sea Research, Part II, 56*(18), 1396–1407. https://doi.org/10.1016/j. dsr2.2008.11.027
- Fay, A. R., & McKinley, G. A. (2013). Global trends in surface ocean pCO₂ from in situ data. Global Biogeochemical Cycles, 27, 541–557. https:// doi.org/10.1002/gbc.20051
- Frankignoulle, M., Canon, C., & Gattuso, J.-P. (1994). Marine calcification as a source of carbon dioxide: Positive feedback of increasing atmospheric CO₂. Limnology and Oceanography, 39(2), 458–462. https://doi.org/10.4319/lo.1994.39.2.0458
- Goddijn-Murphy, L. M., Woolf, D. K., Land, P. E., Shutler, J. D., & Donlon, C. (2015). The OceanFlux Greenhouse Gases methodology for deriving a sea surface climatology of CO₂ fugacity in support of air-sea gas flux studies. *Ocean Science*, *11*(4), 519–541. https://doi.org/10.5194/os-11-519-2015
- Gruber, N. (2009). Fickle trends in the ocean. Nature, 458(7235), 155-156. https://doi.org/10.1038/458155a
- Gruber, N., Gloor, M., Mikaloff Fletcher, S. E., Doney, S. C., Dutkiewicz, S., Follows, M. J., et al. (2009). Oceanic sources, sinks, and transport of atmospheric CO₂. *Global Biogeochemical Cycles*, 23, GB1005. https://doi.org/10.1029/2008GB003349
- Halloran, P. R., Booth, B. B., Jones, C. D., Lambert, F. H., McNeall, D. J., Totterdell, I. J., & Volker, C. (2015). The mechanisms of North Atlantic CO₂ uptake in a large Earth System Model ensemble. *Biogeosciences*, *12*(14), 4497–4508. https://doi.org/10.5194/bg-12-4497-2015
- Henson, S., Lampitt, R., & Johns, D. (2012). Variability in phytoplankton community structure in response to the North Atlantic Oscillation and implications for organic carbon flux. *Limnology and Oceanography*, 57(6), 1591–1601. https://doi.org/10.4319/lo.2012.57.6.1591
- Henson, S. A., Dunne, J. P., & Sarmiento, J. L. (2009). Decadal variability in North Atlantic phytoplankton blooms. Journal of Geophysical Research, 114, C04013. https://doi.org/10.1029/2008JC005139
- Hilligsoe, K. M., Richardson, K., Bendtsen, J., Sorensen, L. L., Nielsen, T. G., & Lyngsgaard, M. M. (2011). Linking phytoplankton community size composition with temperature, plankton food web structure and sea-air CO₂ flux. *Deep Sea Research Part I: Oceanographic Research Papers*, 58(8), 826–838. https://doi.org/10.1016/j.dsr.2011.06.004

Humphreys, M. P., Griffiths, A. M., Achterberg, E. P., Holliday, N. P., Rérolle, V. M. C., Menzel Barraqueta, J.-L., et al. (2016). Multidecadal accumulation of anthropogenic and remineralized dissolved inorganic carbon along the Extended Ellett Line in the Northeast Atlantic Ocean. *Global Biogeochemical Cycles*, 30, 293–310. https://doi.org/10.1002/2015GB005246

Hurrell, J. W., & Deser, C. (2009). North Atlantic climate variability: The role of the North Atlantic Oscillation. Journal of Marine Systems, 78(1), 28–41. https://doi.org/10.1016/j.jmarsys.2008.11.026

Jones, D. C., Ito, T., Takano, Y., & Hsu, W.-C. (2014). Spatial and seasonal variability of the air-sea equilibration timescale of carbon dioxide. Global Biogeochemical Cycles, 28, 1163–1178. https://doi.org/10.1002/2014GB004813

Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., et al. (1996). The NCEP/NCAR 40-year reanalysis project. *Bulletin of the American Meteorological Society*, *77*(3), 437–471. https://doi.org/10.1175/1520-0477(1996)077<0437:TNYRP>2.0.CO;2

Khatiwala, S., Tanhua, T., Mikaloff Fletcher, S., Gerber, M., Doney, S. C., Graven, H. D., et al. (2013). Global ocean storage of anthropogenic carbon. *Biogeosciences*, *10*(4), 2169–2191. https://doi.org/10.5194/bg-10-2169-2013

Klaas, C., & Archer, D. (2002). Association of sinking organic matter with various types of mineral ballast in the deep sea: Implications for the rain ratio. *Global Biogeochemical Cycles*, *16*(4), 1116. https://doi.org/10.1029/2001GB001765

Land, P. E., Shutler, J. D., & Smyth, T. J. (2018). Correction of sensor saturation effects in MODIS oceanic particulate inorganic carbon. IEEE Transactions on Geoscience and Remote Sensing, 56(3), 1466–1474. https://doi.org/10.1109/TGRS.2017.2763456

Lavigne, H., Epitalon, J.-M., & Gattuso, J.-P. (2011). seacarb: Seawater carbonate chemistry with R. R package version 3.0. Retrieved from http://CRAN.R-project.org/package=seacarb

Li, Z., & Cassar, N. (2016). Satellite estimates of net community production based on O₂/Ar observations and comparison to other estimates. Global Biogeochemical Cycles, 30, 735–752. https://doi.org/10.1002/2015GB005314

Longhurst, A. R. (1998). Ecological geography of the sea (p. 397). San Diego: Academic Press.

- Lovenduski, N. S., Gruber, N., Doney, S. C., & Lima, I. D. (2007). Enhanced CO₂ outgassing in the Southern Ocean from a positive phase of the Southern Annular Mode. *Global Biogeochemical Cycles*, *21*, GB2026. https://doi.org/10.1029/2006GB002900
- Marra, J., Ho, C., & Tress, C. (2003). An alternative algorithm for the calculation of primary production from remote sensing data, LDEO Tech. Rep. 2003–1. Palisades, New York: Lamont-Doherty Earth Observatory.
- McKinley, G. A., Fay, A. R., Lovenduski, N., & Pilcher, D. (2017). Natural variability and anthropogenic trends in the ocean carbon sink. Annual Review of Marine Science, 9(1), 125–150. https://doi.org/10.1146/annurev-marine-010816-060529
- Michaels, A. F., & Silver, M. W. (1988). Primary production, sinking fluxes and the microbial food web. Deep Sea Research Part A: Oceanographic Research Papers, 35(4), 473–490. https://doi.org/10.1016/0198-0149(88)90126-4
- Mikaloff Fletcher, S. E., Gruber, N., Jacobson, A. R., Doney, S. C., Dutkiewicz, S., Gerber, M., et al. (2006). Inverse estimates of anthropogenic CO₂ uptake, transport, and storage by the ocean. *Global Biogeochemical Cycles*, *20*, GB2002. https://doi.org/10.1029/2005GB002530

Moore, T. S., Dowell, M. D., & Franz, B. A. (2012). Detection of coccolithophore blooms in ocean color satellite imagery: A generalized approach for use with multiple sensors. *Remote Sensing of Environment*, 117, 249–263. https://doi.org/10.1016/j.rse.2011.10.001

Palevsky, H. I., Ribalet, F., Swalwell, J. E., Cosca, C. E., Cokelet, E. D., Feely, R. A., et al. (2013). The influence of net community production and phytoplankton community structure on CO₂ uptake in the Gulf of Alaska. *Global Biogeochemical Cycles*, 27, 664–676. https://doi.org/ 10.1002/gbc.20058

Pezzulli, S., Stephenson, D. B., & Hannachi, A. (2005). The variability of seasonality. Journal of Climate, 18(1), 71–88. https://doi.org/10.1175/ JCLI-3256.1

- Pickart, R. S., Spall, M. A., Ribergaard, M. H., Moore, G. W. K., & Milliff, R. F. (2003). Deep convection in the Irminger Sea forced by the Greenland tip jet. *Nature*, 424(6945), 152–156. https://doi.org/10.1038/nature01729
- Reynolds, R. W., Smith, T. M., Liu, C., Chelton, D. B., Casey, K. S., & Schlax, M. G. (2007). Daily high-resolution-blended analyses for sea surface temperature. *Journal of Climate*, 20(22), 5473–5496. https://doi.org/10.1175/2007JCL11824.1

Richardson, A. J., Walne, A. W., John, A. W. G., Jonas, T. D., Lindley, J. A., Sims, D. W., et al. (2006). Using Continuous Plankton Recorder data. *Progress in Oceanography*, 68(1), 27–74. https://doi.org/10.1016/j.pocean.2005.09.011

Richardson, T. L., & Jackson, G. A. (2007). Small phytoplankton and carbon export from the surface ocean. Science, 315(5813), 838–840. https://doi.org/10.1126/science.1133471

Sabine, C. L., Feely, R. A., Gruber, N., Key, R. M., Lee, K., Bullister, J. L., et al. (2004). The oceanic sink for anthropogenic CO₂. Science, 305(5682), 367–371. https://doi.org/10.1126/science.1097403

Sabine, C. L., Hankin, S., Koyuk, H., Bakker, D. C. E., Pfeil, B., Olsen, A., et al. (2013). Surface Ocean CO₂ Atlas (SOCAT) gridded data products. *Earth System Science Data*, 5(1), 145–153. https://doi.org/10.5194/essd-5-145-2013

Schuster, U., McKinley, G. A., Bates, N., Chevallier, F., Doney, S. C., Fay, A. R., et al. (2013). An assessment of the Atlantic and Arctic sea-air CO₂ fluxes, 1990-2009. *Biogeosciences*, *10*(1), 607–627. https://doi.org/10.5194/bg-10-607-2013

- Schuster, U., Watson, A. J., Bates, N. R., Corbiere, A., Gonzalez-Davila, M., Metzl, N., et al. (2009). Trends in North Atlantic sea-surface fCO(2) from 1990 to 2006. Deep Sea Research Part II: Topical Studies in Oceanograph, 56(8–10), 620–629. https://doi.org/10.1016/j.dsr2.2008.12.011
- Shiskin, J., Young, A. J., & Musgrave, J. C. (1967). The X-11 variant of the Census Method II Seasonal Adjustment Program, US Dept of Commerce (68 pp.).
- Shutler, J. D., Land, P. E., Brown, C. W., Findlay, H. S., Donlon, C. J., Medland, M., et al. (2013). Coccolithophore surface distributions in the North Atlantic and their modulation of the air-sea flux of CO₂ from 10 years of satellite Earth observation data. *Biogeosciences*, *10*(4), 2699–2709. https://doi.org/10.5194/bg-10-2699-2013
- Shutler, J. D., Land, P. E., Piolle, J., Woolf, D. K., Goddijn-Murphy, L., Paul, F., et al. (2016). FluxEngine: A flexible processing system for calculating atmosphere–ocean carbon dioxide gas fluxes and climatologies. *Journal of Atmospheric and Oceanic Technology*, 33(4), 741–756. https://doi.org/10.1175/JTECH-D-14-00204.1
- Siegel, D. A., Buesseler, K. O., Doney, S. C., Sailley, S. F., Behrenfeld, M. J., & Boyd, P. W. (2014). Global assessment of ocean carbon export by combining satellite observations and food-web models. *Global Biogeochemical Cycles*, 28, 181–196. https://doi.org/10.1002/ 2013GB004743
- Takahashi, T., Olafsson, J., Goddard, J. G., Chipman, D. W., & Sutherland, S. C. (1993). Seasonal variation of CO₂ and nutrients in the highlatitude surface oceans: A comparative study. *Global Biogeochemical Cycles*, 7(4), 843–878. https://doi.org/10.1029/93GB02263
- Takahashi, T., Sutherland, S. C., Sweeney, C., Poisson, A., Metzl, N., Tilbrook, B., et al. (2002). Global sea-air CO₂ flux based on climatological surface ocean pCO(2), and seasonal biological and temperature effects. *Deep Sea Research Part II: Topical Studies in Oceanography*, 49(9–10), 1601–1622. https://doi.org/10.1016/S0967-0645(02)00003-6
- Takahashi, T., Sutherland, S. C., Wanninkhof, R., Sweeney, C., Feely, R. A., Chipman, D. W., et al. (2009). Climatological mean and decadal change in surface ocean pCO(2), and net sea-air CO₂ flux over the global oceans. *Deep Sea Research Part II: Topical Studies in Oceanography*, *56*(8–10), 554–577. https://doi.org/10.1016/j.dsr2.2008.12.009



- Thomas, H., Prowe, A. E. F., Lima, I. D., Doney, S. C., Wanninkhof, R., Greatbatch, R. J., et al. (2008). Changes in the North Atlantic Oscillation influence CO₂ uptake in the North Atlantic over the past 2 decades. *Global Biogeochemical Cycles*, *22*, GB4027. https://doi.org/10.1029/2007GB003167
- Tilstone, G., Xie, Y., Robinson, C., Serret, P., Raitsos, D., Powell, T., et al. (2015). Satellite estimates of net community production indicate predominance of net autotrophy in the Atlantic Ocean. *Remote Sensing of Environment*, *164*, 254–269. https://doi.org/10.1016/j. rse.2015.03.017
- Ullman, D. J., McKinley, G. A., Bennington, V., & Dutkiewicz, S. (2009). Trends in the North Atlantic carbon sink: 1992–2006. *Global Biogeochemical Cycles*, 23, GB4011. https://doi.org/10.1029/2008GB003383
- Vantrepotte, V., & Melin, F. (2011). Inter-annual variations in the SeaWiFS global chlorophyll a concentration (1997–2007). Deep Sea Research Part I: Oceanographic Research Papers, 58(4), 429–441. https://doi.org/10.1016/j.dsr.2011.02.003
- Visbeck, M., Chassignet, E. P., Curry, R. G., Delworth, T. L., Dickson, R. R., & Krahmann, G. (2003). The Ocean's response to North Atlantic Oscillation variability. In J. W. Hurrell, Y. Kushnir, G. Ottersen, & M. Visbeck (Eds.), *The North Atlantic Oscillation: Climatic significance and environmental impact* (pp. 1–35). Washington, DC: American Geophysical Union. https://doi.org/10.1029/134GM06
- Völker, C., Wallace, D. W. R., & Wolf-Gladrow, D. A. (2002). On the role of heat fluxes in the uptake of anthropogenic carbon in the North Atlantic. *Global Biogeochemical Cycles*, 16(4), 1138. https://doi.org/10.1029/2002GB001897
- Watson, A. J., Schuster, U., Bakker, D. C., Bates, N. R., Corbière, A., González-Dávila, M., et al. (2009). Tracking the variable North Atlantic sink for atmospheric CO₂. Science, 326(5958), 1391–1393. https://doi.org/10.1126/science.1177394
- Weiss, R. F. (1974). Carbon dioxide in water and seawater: The solubility of a non-ideal gas. *Marine Chemistry*, 2(3), 203–215. https://doi.org/ 10.1016/0304-4203(74)90015-2
- Westberry, T., Behrenfeld, M. J., Siegel, D. A., & Boss, E. (2008). Carbon-based primary productivity modeling with vertically resolved photoacclimation. *Global Biogeochemical Cycles*, 22, GB2024. https://doi.org/10.1029/2007GB003078
- Woolf, D. K., Land, P. E., Shutler, J., Goddijn-Murphy, L., & Donlon, C. J. (2016). On the calculation of air-sea fluxes of CO₂ in the presence of temperature and salinity gradients. *Journal of Geophysical Research: Oceans, 121*, 1229–1248. https://doi.org/10.1002/2015JC011427
- Yasunaka, S., Nojiri, Y., Nakaoka, S., Ono, T., Whitney, F. A., & Telszewski, M. (2014). Mapping of sea surface nutrients in the North Pacific: Basinwide distribution and seasonal to interannual variability. *Journal of Geophysical Research: Oceans*, 119, 7756–7771. https://doi.org/ 10.1002/2014JC010318
- Zweng, M. M., Reagan, J. R., Antonov, J. I., Locarnini, R. A., Mishonov, A. V., Boyer, T. P., et al. (2013). World Ocean Atlas 2013. In S. Levitus (Ed.), Salinity, NOAA Atlas NESDIS 74 (Vol. 2, p. 39). Silver Spring, MD.