

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

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**Key points:**

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

**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 CO<sub>2</sub>. 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 CO<sub>2</sub> difference ( $\Delta p\text{CO}_2$ ) on seasonal and interannual timescales. We demonstrate that, on seasonal timescales, the subpolar North Atlantic  $\Delta p\text{CO}_2$  signal is predominantly correlated with biological processes, whereas seawater temperature dominates in the subtropics. However, the same factors do not necessarily control  $\Delta p\text{CO}_2$  on interannual timescales. Our results imply that the mechanisms driving seasonal variability in  $\Delta p\text{CO}_2$  cannot necessarily be extrapolated to predict how  $\Delta p\text{CO}_2$ , and thus the North Atlantic CO<sub>2</sub> sink, may respond to increases in anthropogenic CO<sub>2</sub> over longer timescales.

## Plain language summary

As atmospheric carbon dioxide (CO<sub>2</sub>) concentrations rise due to anthropogenic emissions, the ocean is taking up more CO<sub>2</sub>; a process known as the oceanic CO<sub>2</sub> sink. The North Atlantic is a major anthropogenic CO<sub>2</sub> 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<sub>2</sub> sink. Often the factors that affect oceanic uptake of CO<sub>2</sub> 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<sub>2</sub> 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 CO<sub>2</sub> from the atmosphere, taking up approximately one third of emissions since pre-industrial times [Khaliwala *et al.*, 2013]. The high latitude North Atlantic has one of the highest uptake rates of atmospheric CO<sub>2</sub> per square metre [Mikaloff-Fletcher *et al.*, 2006], accounting for 23% of oceanic anthropogenic CO<sub>2</sub> storage, whilst only constituting 15% of the global ocean surface area [Sabine *et al.*, 2004]. However, recent studies suggest that the North Atlantic CO<sub>2</sub> 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<sub>2</sub> 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<sub>2</sub> concentration difference across the boundary layer determines the net direction of CO<sub>2</sub> transfer [Woolf *et al.*, 2016], i.e. the difference between the partial pressure of CO<sub>2</sub> ( $p\text{CO}_2$ ) in seawater and the overlying atmosphere ( $\Delta p\text{CO}_2$ ). 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<sub>2</sub> transfer. Where  $\Delta p\text{CO}_2$  is positive (seawater  $p\text{CO}_2 >$  atmospheric  $p\text{CO}_2$ ), the water is oversaturated, implying a net flux from sea to air, i.e. a potential CO<sub>2</sub> ‘source’. The opposite case, where  $\Delta p\text{CO}_2$  is negative and the ocean is undersaturated, implies a CO<sub>2</sub> ‘sink’. Atmospheric  $p\text{CO}_2$  is homogeneous relative to seawater, so seawater  $p\text{CO}_2$  is typically the dominant control on  $\Delta p\text{CO}_2$  direction. Thus, biogeochemical and hydrographic processes can modify  $\Delta p\text{CO}_2$  if they alter the seawater  $p\text{CO}_2$ . Cooler water has a greater capacity to store dissolved inorganic carbon (DIC) than warm water, as CO<sub>2</sub> solubility is inversely proportional to water temperature. Cooler water reduces seawater  $p\text{CO}_2$ , helping to drive negative  $\Delta p\text{CO}_2$ , while warming has the opposite effect. Net community production (NCP, primary production minus respiration) takes up DIC from the seawater through photosynthesis, decreasing seawater  $p\text{CO}_2$  and contributing to negative  $\Delta p\text{CO}_2$ . Calcification consumes DIC, but is a CO<sub>2</sub> source due to the accompanying net release of CO<sub>2</sub> into the water [Frankignoulle *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 CO<sub>2</sub> sink in the subpolar North Atlantic and a neutral to weak sink in the subtropical North Atlantic [Schuster *et al.*, 2013].

91 An additional biological influence on the air-sea CO<sub>2</sub> flux has been posited:  
92 phytoplankton community structure [Hilligsøe *et al.*, 2011], with functional types such as  
93 diatoms, which are thought to export organic carbon most efficiently, expected to have a  
94 dominant effect [Michaels and Silver, 1988]. However, small phytoplankton have also been  
95 found to influence CO<sub>2</sub> uptake and export [Palevsky *et al.*, 2013; Richardson and Jackson,  
96 2007] and in the North Atlantic dinoflagellate abundance was found to strongly correlate with  
97 organic carbon flux at 2000 m [Henson *et al.*, 2012]. Whilst calcifying phytoplankton (e.g.  
98 coccolithophores) can also modify seawater pCO<sub>2</sub> during formation, they may also contribute  
99 to efficient organic carbon transfer to depth [Klaas and Archer, 2002].

100  
101 The potential controls on the North Atlantic CO<sub>2</sub> sink at different timescales are not  
102 well understood. For example, in a model study Bennington *et al.* [2009] found that  
103 biological activity dominated the seasonal cycle of seawater pCO<sub>2</sub>, but not its interannual  
104 variability. On these longer timescales, the North Atlantic Oscillation (NAO), the dominant  
105 climate variability mode in the region, could affect oceanic CO<sub>2</sub> uptake [Gruber *et al.*, 2009]  
106 and interior CO<sub>2</sub> storage [Humphreys *et al.*, 2016]. In a positive NAO phase, the North  
107 Atlantic Current increases in strength [Visbeck *et al.*, 2003] bringing warm waters with  
108 relatively low DIC concentration into the subpolar northeast Atlantic. Despite the warm  
109 water, the low DIC results in an intensified CO<sub>2</sub> sink in that region, while in the northwest  
110 Atlantic an intensified Labrador Current brings cooler waters with relatively high DIC from  
111 the Arctic, which, despite the cool water, results in a weaker CO<sub>2</sub> sink [Völker *et al.*, 2002].  
112 In the subtropical Atlantic a positive NAO phase has the effect of reducing mixing and  
113 increasing surface water temperatures, which result in lower carbon uptake [Gruber, 2009].

114  
115 Using observational datasets we examine the hypothesis, suggested by a previous  
116 model study [Bennington *et al.*, 2009], that the dominant influences on  $\Delta p\text{CO}_2$  in the North  
117 Atlantic are different at seasonal and interannual timescales. One approach is to separate the  
118 effects using a climatological mass balance technique [Ayers and Lozier, 2012], however to  
119 specifically test the importance of potential mechanisms at different timescales, a method to  
120 decompose a time series into its seasonal and interannual components is needed. Here we  
121 apply a novel decomposition approach to a combination of satellite and *in situ* observations.  
122 We test whether proposed mechanisms for controlling  $\Delta p\text{CO}_2$  are potentially valid on both  
123 seasonal and longer timescales e.g. that biological effects dominate over temperature effects  
124 at high latitudes.

## 2. Methods

Monthly gridded fugacity of seawater CO<sub>2</sub> ( $f\text{CO}_2$ ) for the North Atlantic was downloaded from the SOCAT v3 database (Bakker *et al.*, 2016; [www.socat.info](http://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  $p\text{CO}_2$  was calculated from  $f\text{CO}_2$  using the equations and constants provided in the seacarb R package v3 [Lavigne *et al.*, 2011]. The data were then gridded to a  $1\times 1^\circ$  grid following the SOCAT method [Sabine *et al.*, 2013]. To calculate the  $\Delta p\text{CO}_2$ , atmospheric molar CO<sub>2</sub> concentration was obtained from the NOAA Marine Boundary Layer reference dataset (<https://www.esrl.noaa.gov/gmd/ccgg/mb/index.html>). These were converted to  $p\text{CO}_2(\text{air})$  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  $\mu\text{m}$  (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  $1\times 1^\circ$  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 (CPR) survey (Richardson *et al.*, 2006; dataset doi: [10.7487/2016.194.1.988](https://doi.org/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  $\Delta p\text{CO}_2$  at both seasonal and interannual timescales. Although the CbPM model [Westberry *et al.*,

2008] displays negative correlation between PP and  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  (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  $\Delta p\text{CO}_2$  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, 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,  $\Delta p\text{CO}_2$  was averaged within Longhurst provinces [Longhurst, 1998], as were all satellite-derived and CPR 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 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  $\text{CO}_2$ , and change in  $\text{CO}_2$  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  $\Delta p\text{CO}_2$ , 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  $\Delta p\text{CO}_2$ , 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 non-parametric Spearman correlation coefficient between the different components of  $\Delta p\text{CO}_2$  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 non-parametric partial correlation coefficients to assess association between  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  is correlated with PIC, however PIC is also potentially correlated with NCP which is itself correlated with  $\Delta p\text{CO}_2$ . Partial correlation analysis allows us to determine whether PIC is statistically significantly correlated with  $\Delta p\text{CO}_2$  whilst controlling for the effect of NCP. We also test the correlation between  $\Delta p\text{CO}_2$  and NCP, and between  $\Delta p\text{CO}_2$  and NAO while controlling for SST, and the correlation between  $\Delta p\text{CO}_2$  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  $\text{CO}_2$  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  $\text{CO}_2$  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  $\Delta p\text{CO}_2$ , rather than from a direct correlation between PIC and  $\Delta p\text{CO}_2$ . 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  $\text{CaCO}_3$  during calcification releases  $\sim 0.6$  mole of  $\text{CO}_2$  into the water [Frankignoulle *et al.*, 1994]. On a longer timescale, we expect the export of  $\text{CaCO}_3$  to result in a reduction in surface  $p\text{CO}_2$  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  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  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  $\text{CO}_2$  uptake. The exception is in the northwest Atlantic, where dinoflagellate abundance is positively correlated with  $\Delta p\text{CO}_2$ . 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  $\Delta p\text{CO}_2$  and diatom abundance thus fits this canonical view. However, the negative correlation between the seasonal component of  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  whilst controlling for diatom abundance confirms that dinoflagellate abundance is directly correlated with  $\Delta p\text{CO}_2$  (i.e. the correlation does not arise just because dinoflagellate abundance is correlated with diatom abundance, which itself is correlated with  $\Delta p\text{CO}_2$ ). Our analysis suggests therefore that it is not necessarily the relative abundance of one phytoplankton functional type or another that covaries with  $\Delta p\text{CO}_2$ , 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  $\Delta p\text{CO}_2$  with PP, NCP and PIC.

The NAO is positively correlated with  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$ ). However, in the North Atlantic Drift and Atlantic Arctic provinces, NAO and  $\Delta p\text{CO}_2$  are significantly correlated, even accounting for SST, i.e. positive NAO conditions result in increased  $\Delta p\text{CO}_2$  (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  $\Delta p\text{CO}_2$  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  $\text{CO}_2$  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  $\Delta p\text{CO}_2$ . 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 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  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  response to SST at seasonal scales is no longer evident at interannual timescales. The exception is PP for which negative correlations with  $\Delta p\text{CO}_2$  exist throughout the North Atlantic at all timescales. On interannual timescales, SST is positively correlated, and NCP is negatively correlated, with  $\Delta p\text{CO}_2$  in subpolar regions. Therefore, SST and NCP appear to compete to alter  $\Delta p\text{CO}_2$ . This contrasts with the findings at seasonal scales that imply the temperature effect on  $\Delta p\text{CO}_2$  is secondary to biological effects. The differences in spatial patterns between Figures 2 and 3 suggest that the processes affecting  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  in the North Atlantic Drift Province; however on interannual timescales, NAO is negatively correlated with  $\Delta p\text{CO}_2$  in the same region. How can this apparent contradiction be reconciled? The answer may lie in the different timescales on which the mechanisms affecting  $\Delta p\text{CO}_2$  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 timescale, acts to reduce ocean uptake. However, at the interannual scale, positive NAO periods are associated with increased ocean carbon uptake (decreased  $\Delta p\text{CO}_2$ ) 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  $\Delta p\text{CO}_2$  over different time scales is clearly shown in our analysis. The decomposition method used here therefore allows novel insights into how the factors controlling  $\Delta p\text{CO}_2$  may depend on the timescale under consideration.

The equilibration timescale of  $\text{CO}_2$  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  $\Delta p\text{CO}_2$  signal established by seasonal biological or temperature variability. Halloran *et al.* [2015] identify four mechanisms hypothesised to control variability in ocean  $\text{CO}_2$  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  $\Delta p\text{CO}_2$ . 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  $\Delta p\text{CO}_2$  by separating seasonal and interannual timescales. On seasonal timescales, 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 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  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$ .

Our analysis shows that understanding the mechanisms underlying seasonal variability in  $\Delta p\text{CO}_2$  does not directly inform on how the North Atlantic  $\text{CO}_2$  sink responds to interannual forcing. Mechanistic understanding of the North Atlantic  $\text{CO}_2$  sink should not therefore be based solely on seasonal drivers, but should also consider interannual variability. At decadal timescales the processes affecting  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  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 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  $\Delta p\text{CO}_2$  variability [Breedon 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  $\text{CO}_2$ .

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**Figure Captions**

**Figure 1:** Relative importance of temperature and net biological effects on climatological  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  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  $\Delta p\text{CO}_2$  against potential controls.

Figure 1.

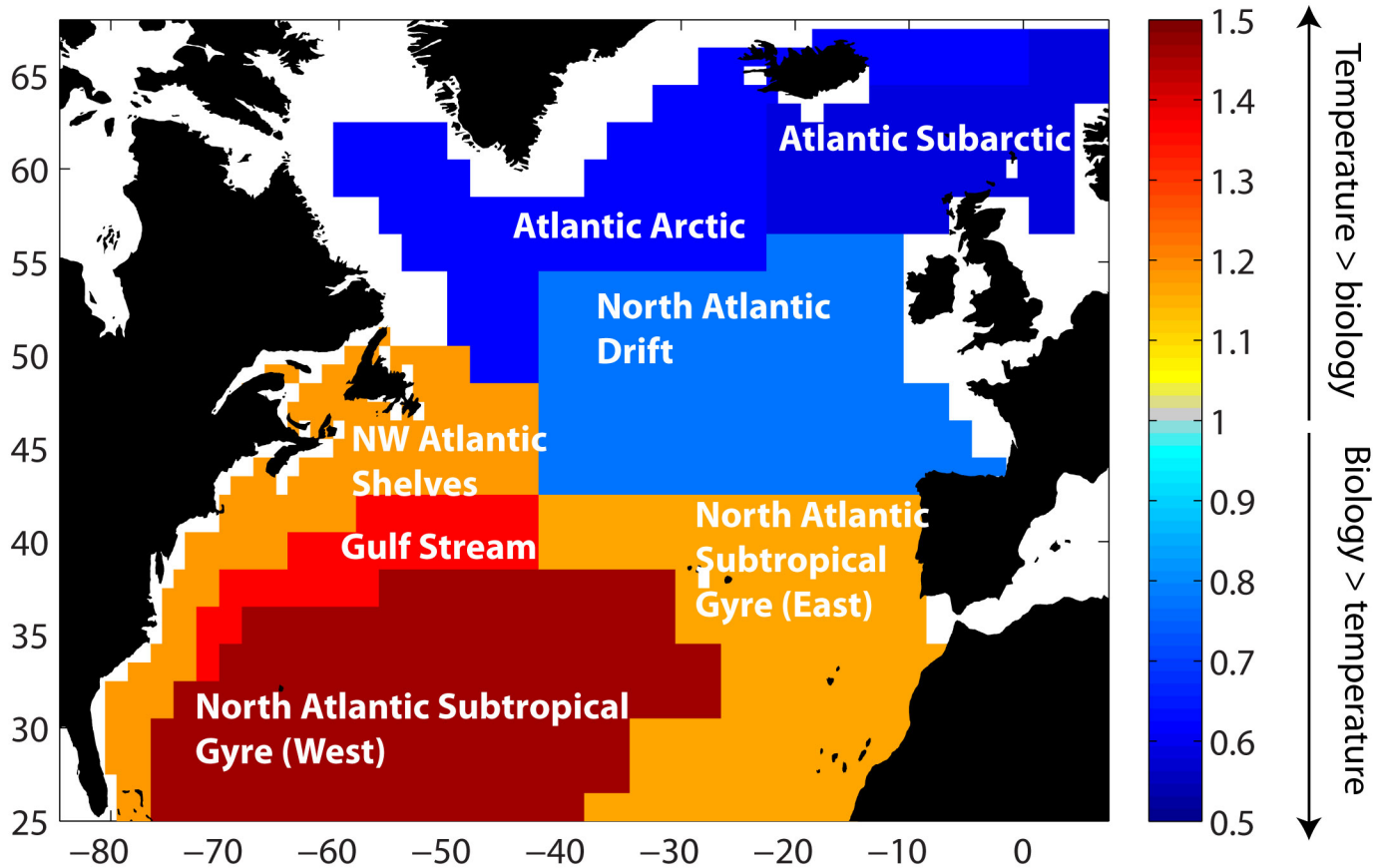


Figure 2.



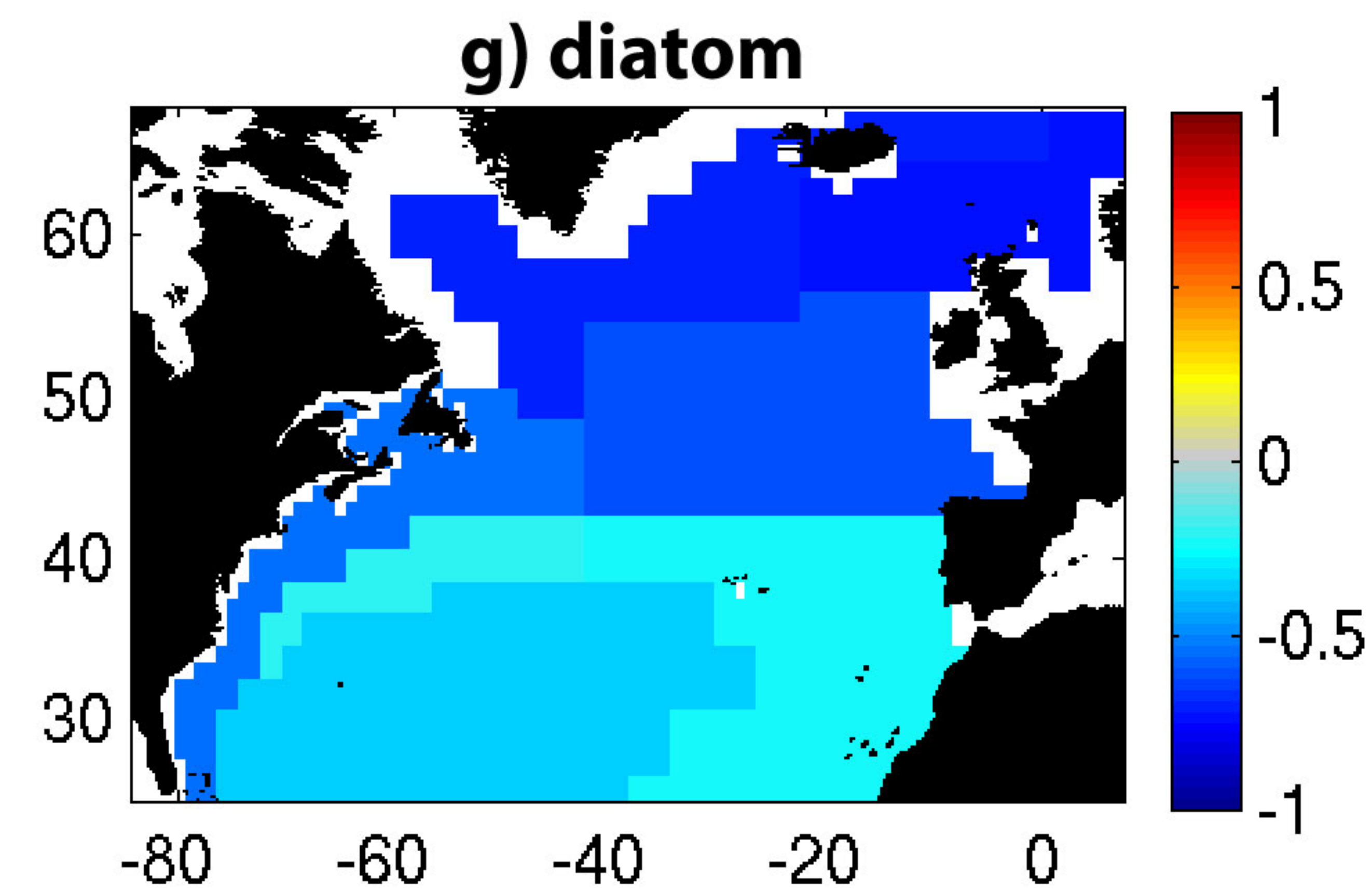
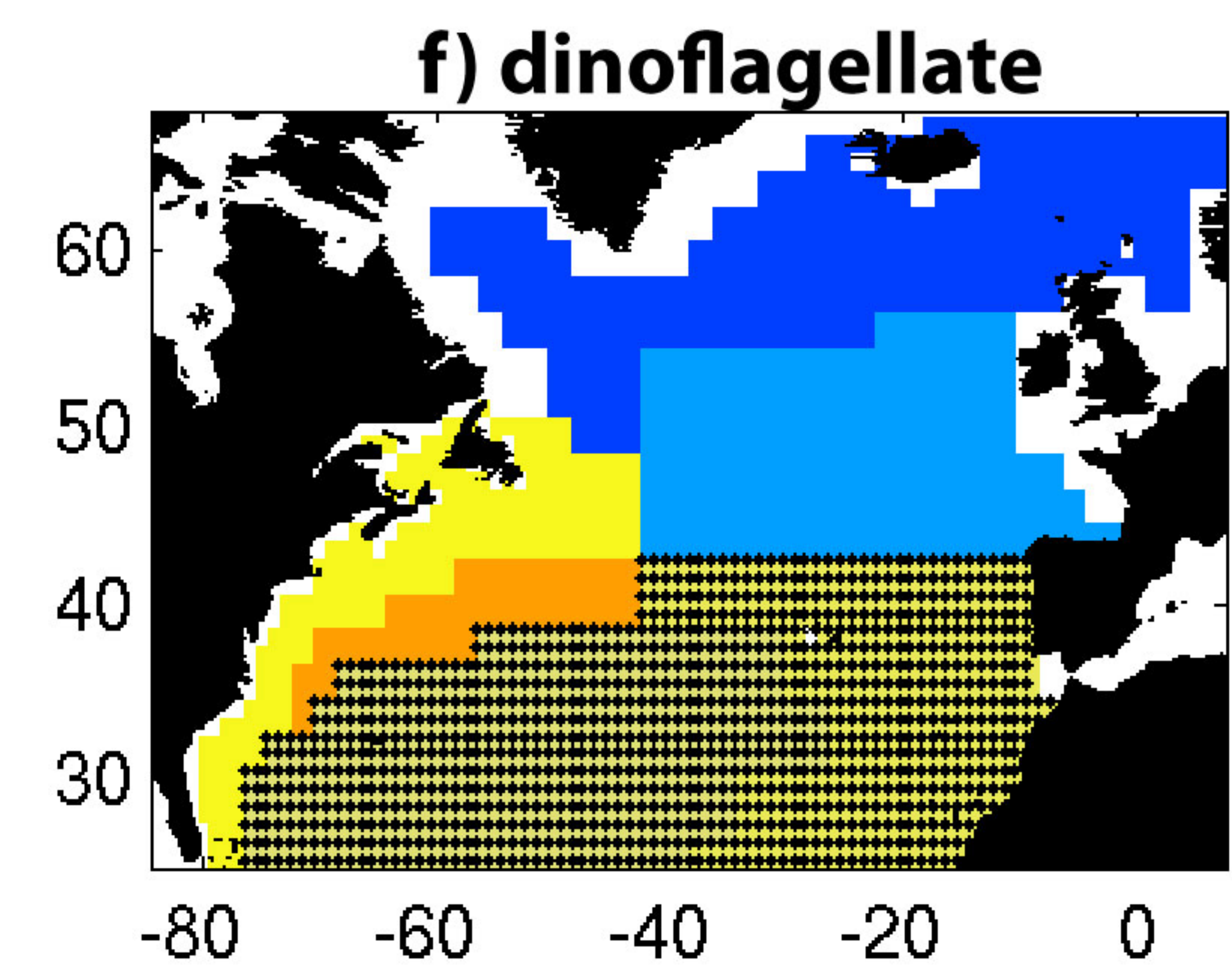
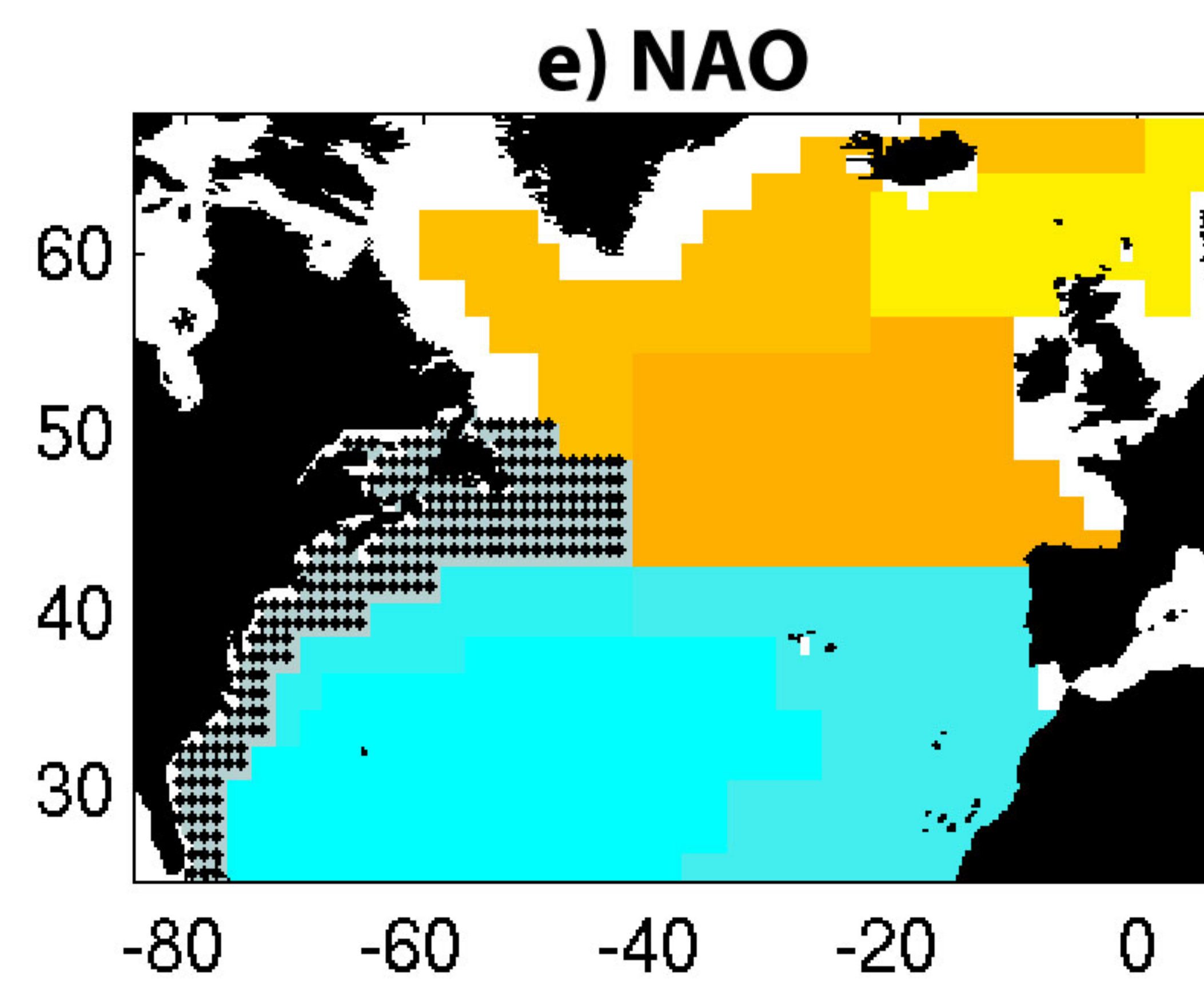
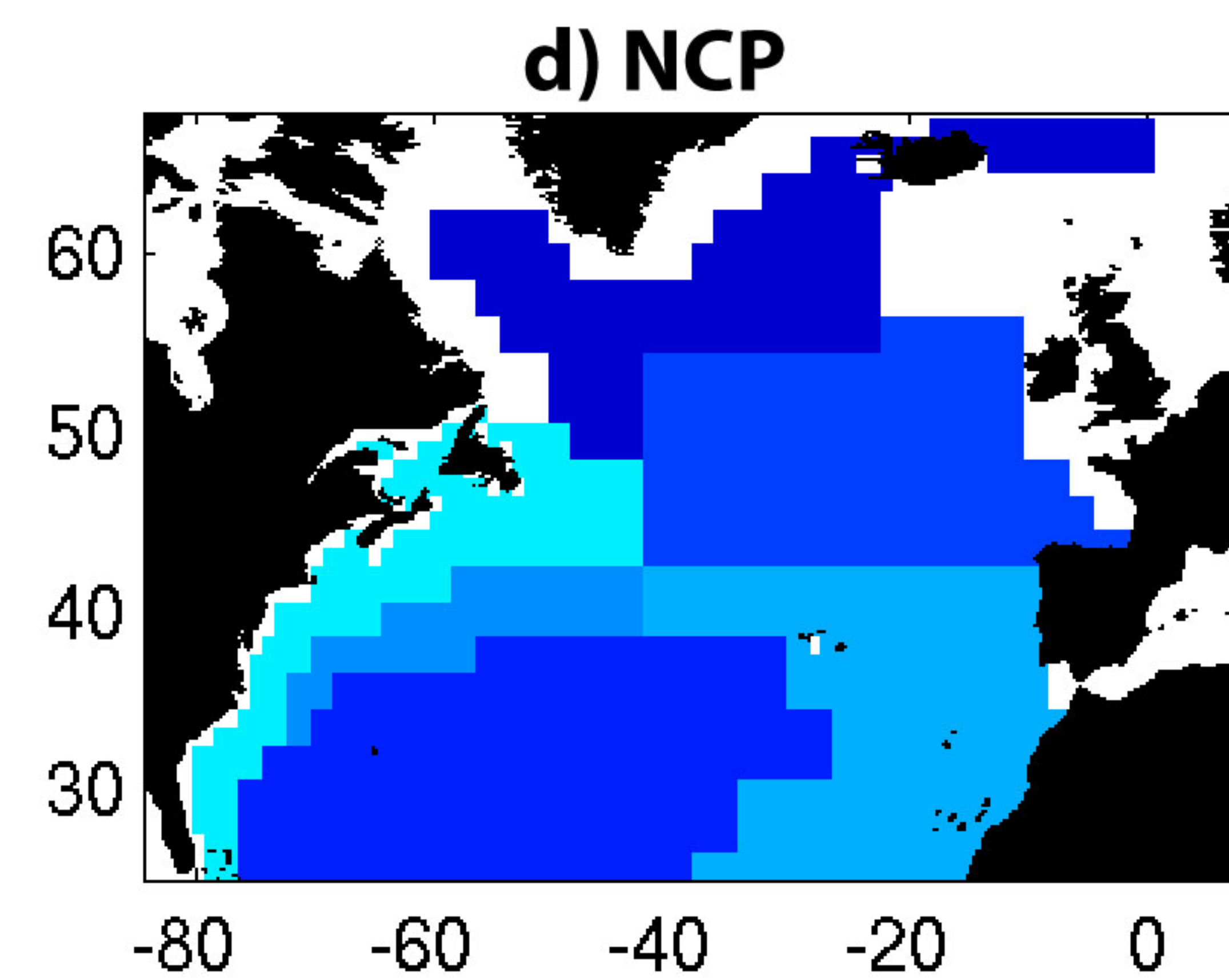
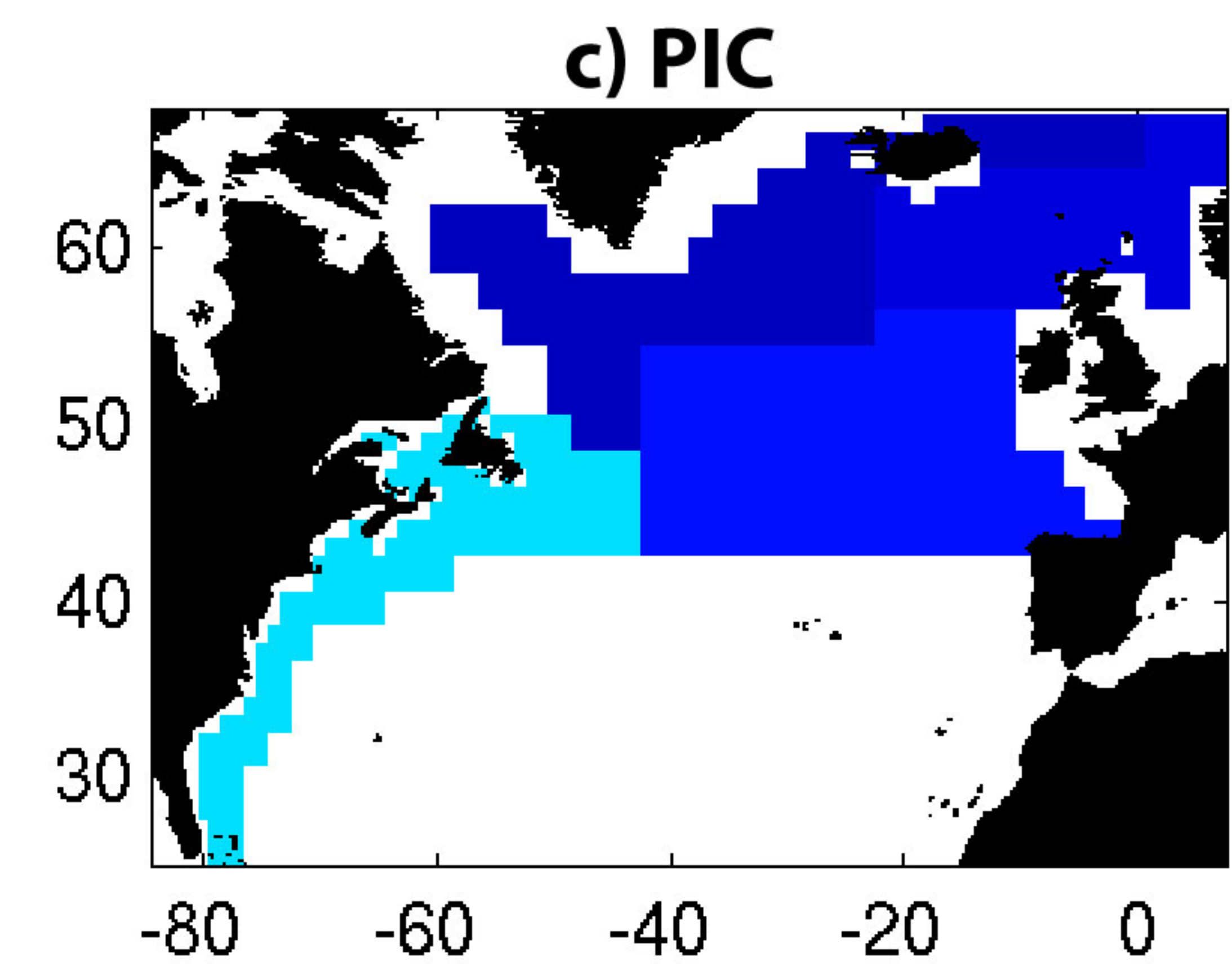
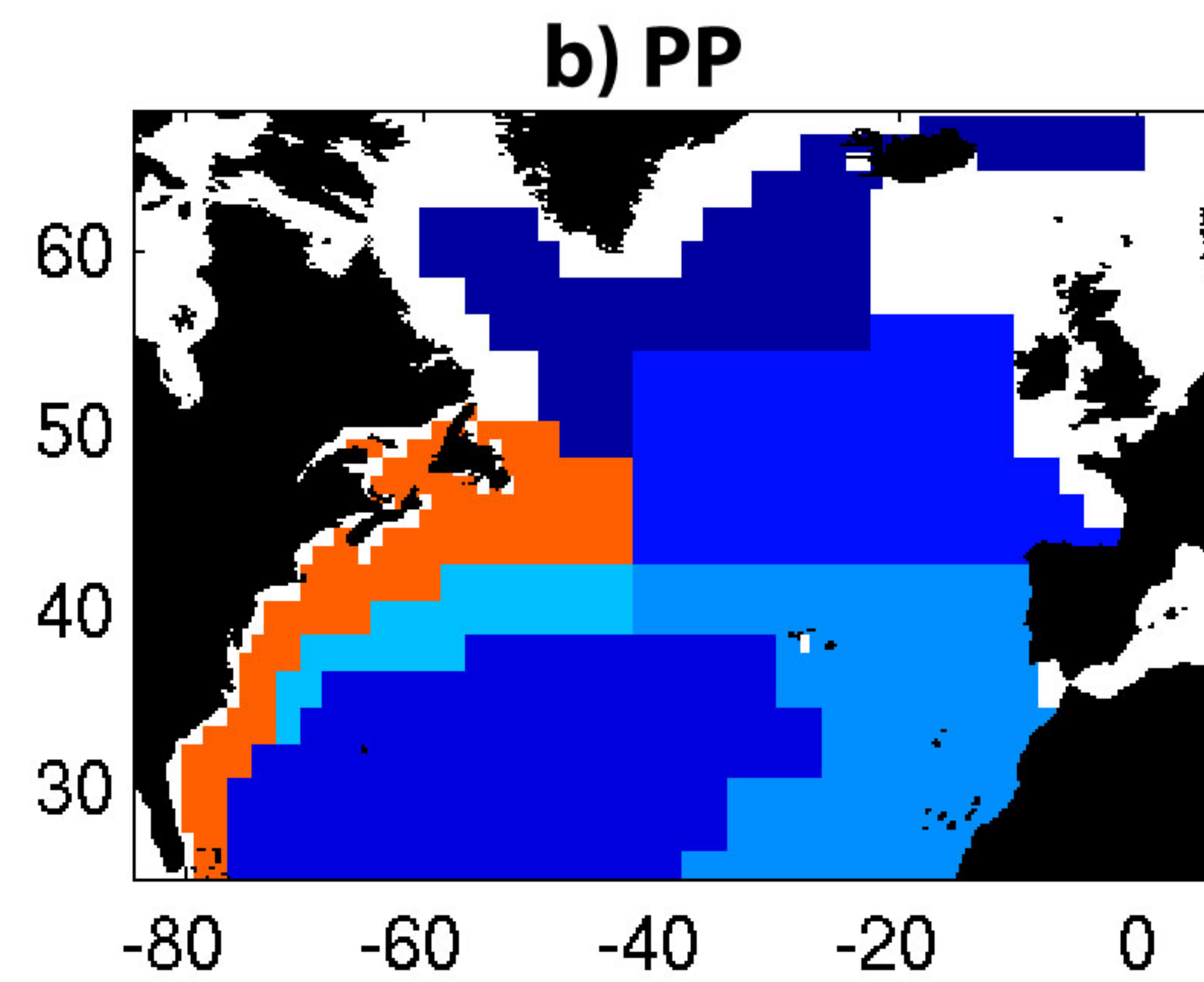
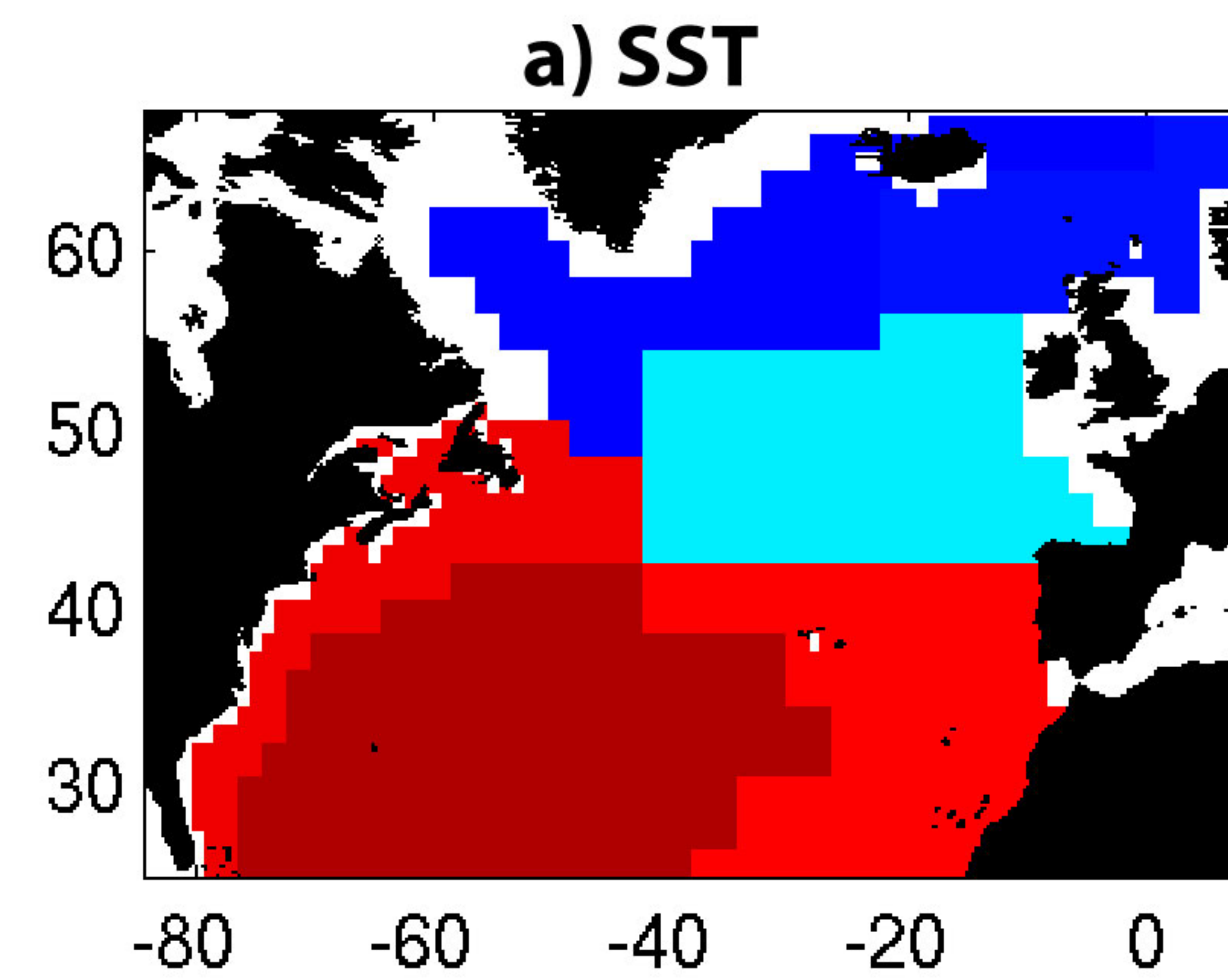




Figure 3.

