



RESEARCH ARTICLE

10.1029/2018GB006132

Key Points:

- Surface nitrate and phosphate concentrations have been higher after 2011 in the North Atlantic Subtropical Gyre
- The winter seasons of 2010 and 2011 had anomalously high concentrations of surface nitrate across the North Atlantic
- This caused an up to fourfold increase in new production in the Central North Atlantic

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Citation:

Macovei, V. A., Torres-Valdés, S., Hartman, S. E., Schuster, U., Moore, C. M., Brown, P. J., et al. (2019). Temporal variability in the nutrient biogeochemistry of the surface north Atlantic: 15 years of ship of opportunity data. *Global Biogeochemical Cycles*, 33. <https://doi.org/10.1029/2018GB006132>

Received 14 NOV 2018

Accepted 14 OCT 2019

Accepted article online 31 OCT 2019

Temporal Variability in the Nutrient Biogeochemistry of the Surface North Atlantic: 15 Years of Ship of Opportunity Data

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Abstract Ocean biological processes play an important role in the global carbon cycle via the production of organic matter and its subsequent export. Often, this flux is assumed to be in steady state; however, it is dependent on nutrients introduced to surface waters via multiple mechanisms, some of which are likely to exhibit both intra-annual and interannual variability leading to comparable variability in ocean carbon uptake. Here we test this variability using surface (5 m) inorganic nutrient concentrations from voluntary observing ships and satellite-derived estimates of chlorophyll and net primary production. At lower latitudes, the seasonality is small, and the monthly averages of nitrate:phosphate are lower than the canonical 16:1 Redfield ratio, implying nitrogen limitation, a situation confirmed via a series of nutrient limitation experiments conducted between Bermuda and Puerto Rico. The nutrient seasonal cycle is more pronounced at higher latitudes, with clear interannual variability. Over a large area of the midlatitude North Atlantic, the winters of 2009/2010 and 2010/2011 had nitrate values more than $1\mu\text{mol L}^{-1}$ higher than the 2002–2017 average, suggesting that during this period, the system may have shifted to phosphorus limitation. This nitrate increase meant that, in the region between 31° and 39° N, new production calculated from nitrate uptake was 20.5g C m^{-2} in 2010, more than four times higher than the median value of the whole observing period. Overall, we suggest that substantial variability in nutrient concentrations and biological carbon uptake occurs in the North Atlantic with interannual variability apparent over a number of different time scales.

1. Introduction

The productivity of the North Atlantic is largely controlled by the supply of nutrients to the upper ocean. Much of the North Atlantic is covered by the North Atlantic Subtropical Gyre (NASTG), an oligotrophic region with surface inorganic nutrient concentrations $< 0.4\mu\text{mol L}^{-1}$ for NO_3^- (nitrate) and $< 0.05\mu\text{mol L}^{-1}$ for PO_4^{3-} (phosphate) (Torres-Valdés et al., 2009). This leads to low levels of net primary productivity (NPP). However, the large surface area and continued production throughout the year makes the integrated net production a significant component of the ocean carbon pump. It is estimated that half of the global primary production of 50Pg year^{-1} occurs in the subtropical gyres (Duce et al., 2008; Emerson et al., 1997). The new production estimates in this area, determined by sediment trap and algorithms applied to remotely sensed chlorophyll, are approximately $1\text{mol C}\cdot\text{m}^{-2}\cdot\text{year}^{-1}$ (Williams & Follows, 1998).

While the subtropics show little variability in the annual cycle of primary production, the midlatitudes are characterized by strong seasonal cycles, with notable peaks during the spring blooms which occur when a deep mixed layer, replenished with nutrients over the winter, stratifies and light limitation is eliminated. This process, and the associated lower levels of production thereafter supported by other nutrient sources, contributes significantly to CO_2 drawdown (Lauderdale et al., 2016). Nevertheless, export production fueled by nutrient supply to the surface ocean is not necessarily a net sink for atmospheric CO_2 , with storage of carbon by the biological pump ultimately determined by the overall use efficiency of nutrients resupplied to the surface (Ito & Follows, 2013), which in turn can be related to the regional sequestration efficiency

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(DeVries et al., 2012). High-resolution nutrient concentration measurements provide a potential method for investigating the time-varying components of these processes.

The nutrients that support primary production are brought to the surface by a series of mechanisms including horizontal advection from neighboring regions (Williams & Follows, 1998), through nutrient streams such as the Gulf Stream-North Atlantic Current (Palter & Lozier, 2008; Pelegrí et al., 1996; Williams et al., 2011), vertical transfer by convective mixing (Mahaffey et al., 2004), and mesoscale eddies (McGillicuddy et al., 1998). Traditionally, it is often assumed that the biological pump operates in quasi steady state—however, many of the terms which ultimately set the size of the biological signal in an oligotrophic region are likely to be sensitive to elements of the ocean circulation, which are clearly not in steady state. For example, the Atlantic Meridional Overturning Circulation (AMOC), an important component of the ocean circulation, is argued to have decreased in intensity through interpretation of water transport across 26° N (Sanchez-Franks & Zhang, 2015; Smeed et al., 2014). Other studies however suggest that this is just a part of natural multidecadal variability since the current indices do not extend over a long enough period to discern a trend (Parker & Ollier, 2016). If the circulation is changing and therefore altering the supply of nutrients to the sea surface, productivity and potentially the efficiency of the biological pump might also be expected to change (Morris & Sanders, 2011).

In this contribution, we investigate biogeochemical variability using diverse data streams. One key feature of suitable data sets is that they should be sustained over many years. For the last few decades, a global network of fixed-point observatories has been developed, producing important time series of biogeochemical variables (Cianca et al., 2007, 2012; Neuer et al., 2007; Steinberg et al., 2001; Sweeney et al., 2003). However, the estimated footprint sizes of these observatories only cover 9–15% of the total ocean surface (Henson et al., 2016). Better spatial coverage is obtained using ship-based observations; however, the temporal resolution of such surveys is low. A solution to this issue is using Ships of Opportunity (SOO), commercial vessels that voluntarily collect samples for subsequent scientific analysis.

Here we present the findings from the first 15 years of an ongoing time series of SOO data on inorganic nutrient concentrations from the North Atlantic. The objectives of this study are to evaluate surface nutrient variability, investigate potential causes for the observed changes, and evaluate what contribution they might have for the upper ocean productivity. We investigate the evolution of sea surface macronutrient (nitrate + nitrite, phosphate, and silicate) concentrations in recent years and the mechanisms supplying these nutrients to the North Atlantic. Given the observed variability in nutrient availability, we further investigate whether shifts in the nutrient limitation regime of the study area have occurred. We also calculate the new production sustained by the nutrients and compare it to satellite-derived estimates of net primary production. The ship tracks spatial coverage spans between the English Channel, as far south as 14° N and as far west as 72° W, a carbon sink area where the biological component is significant (Takahashi et al., 2009).

2. Materials and Methods

2.1. Sample Collection and Analysis

Samples were collected from a pumped seawater system by crew members onboard three ships chartered by *Geest Line Ltd.* (Fareham, UK): *MV Santa Maria*, *MV Santa Lucia*, and *MV Benguela Stream*, reefers that transit between the United Kingdom and the Caribbean every month (Hartman et al., 2010). The seawater intake is located 3–5 m below sea level depending on the cargo and fuel load (Schuster & Watson, 2007), and for the purpose of this paper, we identify the samples as taken from the surface mixed layer. Water samples were collected every 4 hr in 15ml plastic tubes and frozen immediately for later analysis in the laboratory on shore. It is standard practice to collect open-ocean samples without filtering as long as care is taken not to contaminate the tubes (Hydes et al., 2010). Between 90 and 100 samples were collected during every return voyage.

Samples were analyzed for inorganic macronutrients: nitrate + nitrite (hereafter nitrate), phosphate, and silicate using auto-analyzers following the colorimetric method described by Grasshoff et al. (1999). The auto-analyzers used are mentioned in Table 1. The detection limits computed by SEAL Analytical's own software were on average $0.02\mu\text{mol L}^{-1}$ for NO_3^- and Si(OH)_4 and $0.01\mu\text{mol L}^{-1}$ for PO_4^{3-} . This uses the precision of the analysis when measuring a minimum of 10 consecutive repeats of the lowest concentration calibrant. The precision of the method was assessed using the duplicate measurements of the calibration standards. The standard deviation of the duplicates, expressed as a percentage of the mean, is lower than 2% in 93%,

Table 1
List of the Different Auto-Analyzers Used During This Study

Period	Instrument
2002–2008	Skalar San ++ Continuous Flow Analyzer
2008–2012	SEAL Analytical QuAAtro Autoanalyzer
2012–2017	SEAL Analytical QuAAtro and AA3 HR Continuous Segmented Flow Analyzer

93%, and 92% of cases and lower than 1% in 84%, 87%, and 88% of cases when this was tested ($n = 130$) for nitrate, silicate, and phosphate, respectively. Since 2016, the accuracy of the analytical measurements was tested by analyzing reference materials for the analysis of nutrients in seawater from The General Environmental Technos Co., Ltd., (KANSO) (Aoyama, 2018). Correction factors were applied to the sample results in order to match the certified inorganic nutrient concentrations of the reference materials for the analysis of nutrients in seawater.

2.2. Quality Control

Samples collected near the shore were discarded from the analysis to minimize riverine and shelf influence. Then, in order to eliminate samples that might have been contaminated, several data processing filters were used. Some phosphate concentrations were higher than $1\mu\text{mol L}^{-1}$, which is uncommon in surface waters. It is likely in those cases, contamination occurred during the collection, storage, or transport stages. Samples with phosphate values higher than $1\mu\text{mol L}^{-1}$ and their corresponding values for the other nutrients were discarded from the analysis. Next, the N^* (excess nitrate) was calculated as

$$N^* = [\text{NO}_3^-] - 16 \times [\text{PO}_4^{3-}] \quad (1)$$

(Gruber & Sarmiento, 1997). A compilation of 3,390 data points from the GLODAP v2 surface nutrient values in the subtropical and midlatitude North Atlantic revealed a mean N^* value of -0.81 ± 1.48 (Olsen et al., 2016). Most of the results of this study clustered around the 0 value, but a few isolated points were classed as outliers, and an additional filter applied was eliminating samples with $N^* > 5$ or $N^* < -5$. Following quality control, 13,210 measurements for nitrate, 13,735 for silicate, and 12,264 for phosphate were deemed acceptable between May 2002 and October 2017.

Clearly, overall data quality controls within volunteer observing programs remain a greater challenge than for standard oceanographic observational studies. Consequently, we acknowledge that caution should be applied in the interpretation of observed variability, particularly within lower concentration measurements where difficult to monitor low-level contamination and long-term freezing of samples might both have the potential to introduce subsequent error. The standard errors of the nutrient concentrations in an average year are small (section 3.1) which gives a high signal-to-noise ratio in the time series. The rigorous quality control procedure mitigates against there being a systematic bias in our data. In addition, we compare our data to other nutrient data from the region in order to determine whether the features we observed were consistent with those seen in other data sets obtained through analysis on different instruments by different operators (section 3.2).

2.3. Biogeographical Regions

The ships' tracks cross regions with contrasting nutrient concentrations. For example, while the nitrate values sometimes exceed $10\mu\text{mol L}^{-1}$ near the U.K. coast, they never reach $1\mu\text{mol L}^{-1}$ in the NASTG. We split our area of research into five regions following Ostle et al. (2015). They used a method similar to Hooker et al. (2000) where the second derivatives of in situ temperature, density, and satellite-derived chlorophyll along the ship track were normalized and averaged and then plotted against latitude. This manner of plotting produced four peaks, which were used to define the latitudinal boundaries and split the ship tracks into five regions which broadly match biogeographical regions in the North Atlantic as defined by Longhurst (2006). We adopt the Ostle et al. (2015) selection of regions given that the data assessed here belong to the same research program and were collected simultaneously onboard the same vessels. A map of the ship's crossings with the location of the samples and the boxes that define the five chosen regions is shown in Figure 1, overlain on a satellite chlorophyll map.

2.4. Data Processing and External Data

In order to investigate the seasonal variation of surface nutrients in each region, we first computed monthly means throughout the time series (equation (2)). Next, to study trends or anomalous events, we calculated

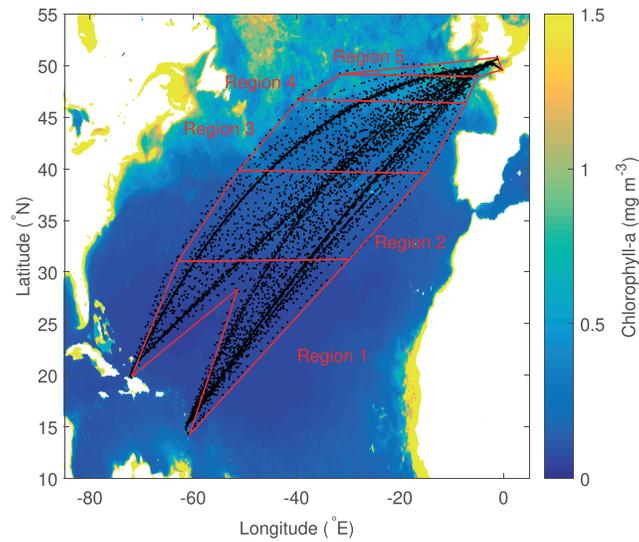


Figure 1. Map of the study area showing all sampling locations in black dots, overlain on a composite satellite-derived chlorophyll-*a* concentration map for the Year 2017 with the upper color axis limit set to 1.5 mg m^{-3} . The separation between the regions is based on the latitudinal boundaries defined by Ostle et al. (2015). The polygons show the geographical extent for which external data sources such as satellite chlorophyll-*a* were assigned to be part of a particular region.

the anomalies by subtracting the long-term mean seasonal cycle from the monthly means (equation (3)). Negative (positive) values indicate that the region's mean nutrient concentration was lower (higher) than expected from the long-term mean seasonal cycle. Furthermore, removing seasonality helps to deal with the autocorrelation that is often found in environmental time series data (Bence, 1995).

$$\text{Average concentration}_x = \frac{\sum_{2002}^{2017} \text{Nutrient concentration}_x}{2017 - 2002 + 1} \quad (2)$$

where x is the month and the calculation is performed for each month between January and December to obtain multiyear averages

$$\text{Nutrient anomaly}_{x,y} = \text{Nutrient concentration}_{x,y} - \text{Average concentration}_x \quad (3)$$

where y is the year.

Alongside our nutrient data, we used several independent data sources to aid our interpretation. Mixed layer depth (MLD) was calculated from Argo data (Holte et al., 2016) as the depth at which the density increases from the surface density by 0.03 kg m^{-3} (de Boyer Montégut et al., 2004). Satellite-derived chlorophyll-*a* was obtained from the MODIS-Aqua satellite database (NASA Goddard Space Flight Center, 2016). Satellite-derived primary production was downloaded from the Ocean Productivity website (Ocean Productivity, 2016) calculated according to Behrenfeld and Falkowski (1997). These data were averaged, seasonally detrended in the same way as the nutrient data and used to determine the biological responses to the effects of variation in nutrient concentrations.

In order to get an estimate of ocean circulation and atmospheric variability, we considered the AMOC and the NAO, respectively. AMOC data, in particular the meridional overturning component transport as measured at 26.5° N , were downloaded from the RAPID website (Smeed et al., 2016). NAO data were downloaded from the National Center for Atmospheric Research website (Hurrell, 2018). These two data sets have not been averaged spatially, with the AMOC transport representing a single zonal transect, whereas the principal component NAO indices are the time series of the leading empirical orthogonal function of sea level pressure anomalies over the Atlantic sector.

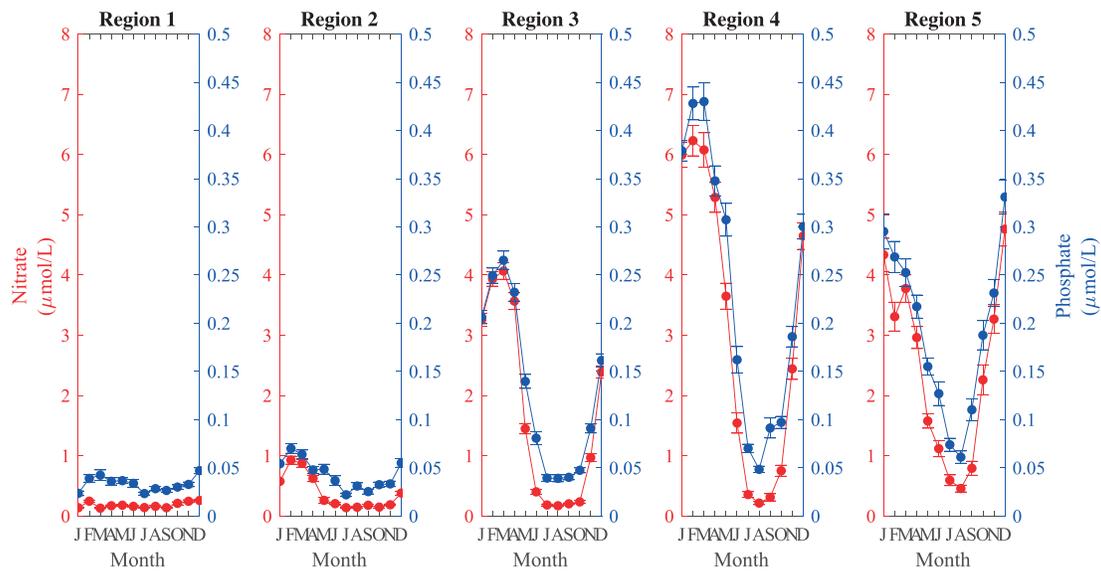


Figure 2. Scatter plots showing the long-term mean seasonal cycle of nitrate (red symbols) and phosphate (blue symbols) values versus month for an average year in the five regions described in Figure 1. The error bars show the standard error of the mean. The vertical axes limits are set in a 16 to 1 nitrate to phosphate ratio.

2.5. Nutrient Limitation Experiment

A series of shipboard nutrient limitation experiments was performed during the fall 2017 Bermuda Atlantic Time Series (BATS) Validation Cruise between Bermuda and Puerto Rico on a meridional transect at approximately 65° W. The experiments were performed three times (“Experiment 1” at 30° N, “Experiment 2” at 24° N, and “Experiment 3” at 20° N). Water was collected before dawn from 15m depth. Transparent Nalgene bottles were filled and placed into an on-deck incubator equipped with blue filters to decrease incoming radiation levels. Triplicate bottles for the control (no nutrient added), nitrate addition (concentration amended to $1\mu\text{mol L}^{-1}$ nitrate), phosphate addition (concentration amended to $0.2\mu\text{mol L}^{-1}$ phosphate), and nitrate + phosphate addition experiments were kept in the incubator for 48 hr, and afterward, 500ml were filtered through GF/F grade glass microfiber filters. Chlorophyll-*a* and phaeophytin were determined by acetone pigment extraction and fluorometer measurement. Unfortunately, we did not have the capacity for trace metal clean sampling within the experiments performed on the cruise. However, previous work within the region (Browning et al., 2017; Moore et al., 2006, 2008) performed using trace metal clean techniques and with and without the addition of iron indicates that bulk community production and biomass accumulation (chlorophyll) are insensitive to iron availability within the region.

3. Results and Discussion

3.1. Long-Term Observations and the Annual Cycle

The time series demonstrates variability in surface nutrient concentrations over a range of spatial and temporal scales. Overall, concentrations displayed the expected latitudinal gradient from high values in the north to lower values within the gyre waters to the south, clearly visible in the annual cycles in Figure 2. Marked seasonality is apparent throughout the majority of the sampled region. While at the lowermost latitudes, nutrient concentrations are relatively constant throughout the year, a more pronounced seasonal cycle in nutrient concentration is observed between 28° and 38° N (Figure 3) implying that convection is a source of nitrate into the photic zone and hence fuels new production. This finding is consistent with the suggestion that wintertime mixing is the main supply mechanism for nutrients in the eastern part of the NASTG, at the ESTOC fixed-point observatory (Cianca et al., 2007). The larger intra-annual variability at latitudes higher than 40° N is expected since this is a region of stronger winter mixing and significant biological activity which determine the annual balance between input and usage of nutrients (Henson et al., 2003; Williams et al., 2000).

The mean seasonal cycles of nitrate and phosphate for each region are shown in Figure 2, with the vertical axes set to the canonical 16:1 (Redfield, 1934) ratio. The lowermost line on each graph defines the nutrient that has the potential of becoming limiting to growth if nitrate and phosphate are consumed in Redfield

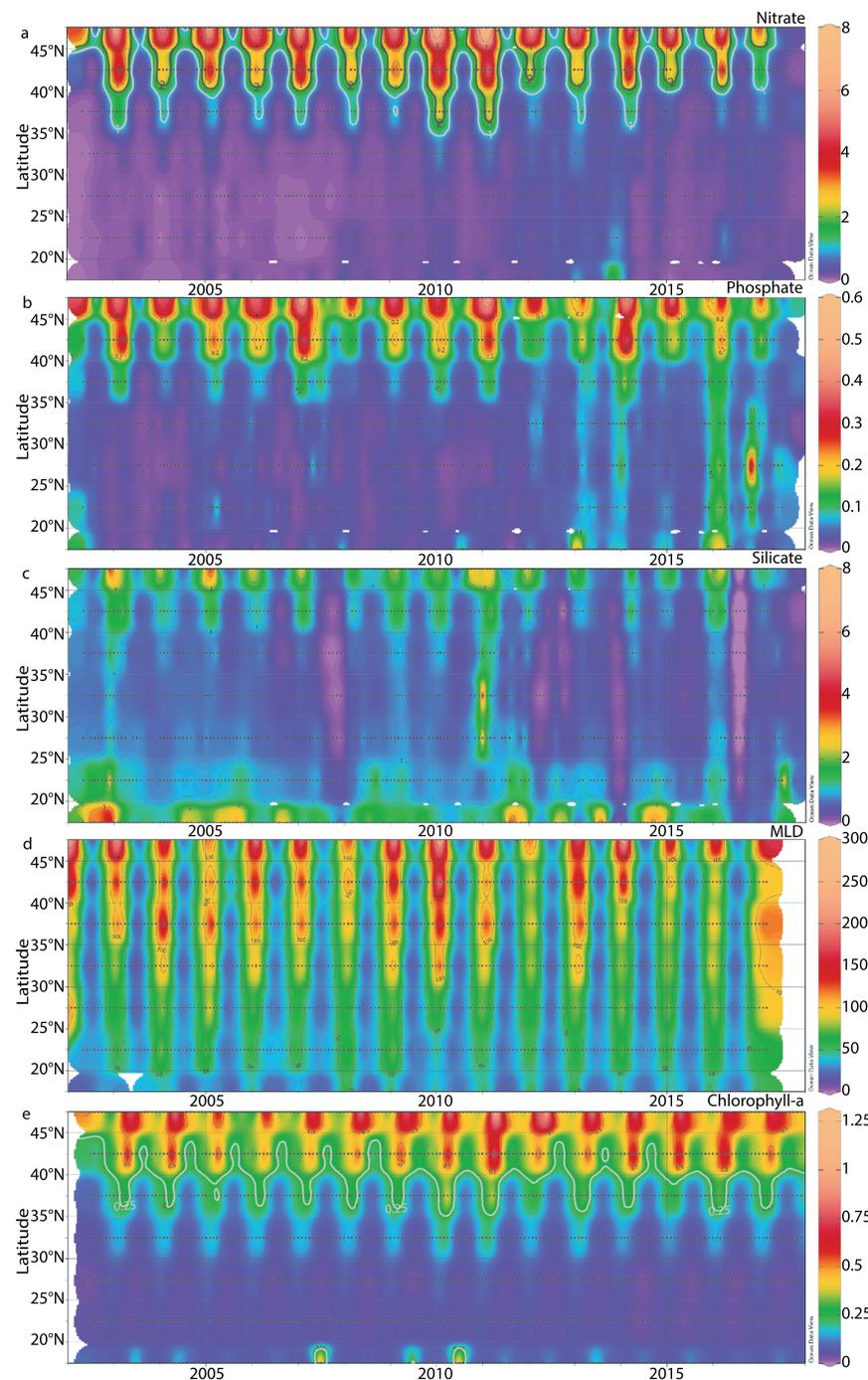


Figure 3. Hovmöller diagrams showing the geographical and temporal distribution of (a) nitrate, (b) phosphate, (c) silicate (all expressed as $\mu\text{mol L}^{-1}$), (d) mixed layer depth (m), and (e) satellite chlorophyll-*a* (mg m^{-3}). Data were averaged every month in 5° latitudinal bands across the study area. The $2\mu\text{mol L}^{-1}$ nitrate contour is emphasized in dark gray. The $1\mu\text{mol L}^{-1}$ nitrate and 0.25mg m^{-3} chlorophyll-*a* contours are emphasized in light gray.

stoichiometry (Neill, 2005). The blue line plotting above the red line indicates N:P ratios below the canonical Redfield ratio.

As expected, the nutrient concentrations increase northward through the regions, particularly in winter. The maximum values range from less than 0.2 and $0.05\mu\text{mol L}^{-1}$ in Region 1 to over 6 and $0.4\mu\text{mol L}^{-1}$ in Region 4 for nitrate and phosphate, respectively. The highest average concentrations are not seen in Region 5

probably because the samples immediately near the shore were discarded from the analysis to avoid riverine influence. Apart from Region 1 where the seasonal variation is very small, the maximum average values are recorded in the winter months, typical of convective mixing being the main source of replenishing nutrients to the surface ocean. The minimum average concentration for both nitrate and phosphate is recorded in July in Region 2 and in August in Regions 3–5.

In Region 1, nitrate is the most deficient nutrient throughout the year, which fits past observations that found nitrate limitation in the NASTG (Meyer et al., 2016; Moore et al., 2013). In Region 3, data suggest that nitrate and phosphate are supplied roughly in a 16:1 ratio during winter, but nitrate is consumed at a faster rate than this ratio in the productive season, which leads to nitrate deficiency in the summer and autumn as shown by the red line plotting below the blue one in Figure 2. Similarly, in Regions 4 and 5, summer nitrate deficiency is observed.

To investigate spatial and temporal variability together, nutrient concentrations were plotted as a Hovmöller diagram. The $2\mu\text{mol L}^{-1}$ nitrate contour was consistently found north of 37.5°N apart from 2010 and 2011 (Figure 3a). Additionally, an increase in sea surface nitrate concentrations occurred in the southern regions during the 2012–2014 and 2016–2017 intervals. The phosphate plot shows features similar to the nitrate one, but in the southern part, the concentrations increase more in the last 3 years of the time series compared to the earlier onset of the increase in nitrate. The silicate plot shows a similar seasonal cycle to the nitrate one in the northern half of the study area, but the amplitude of the variation is smaller (only higher than $3\mu\text{mol L}^{-1}$ in 2 years of the time series). High values are however measured south of 20°N . This is likely a result of intrusions of low-salinity water coming in from the Amazon-Orinoco plume that sometimes reaches the Lesser Antilles and forms the Caribbean Surface Water (Corredor & Morell, 2001). The fact that nitrate and phosphate are not elevated in the Caribbean Surface Water in spite of high concentrations present at the point source (Sharples et al., 2017) suggests that they are consumed en route in the tropical Atlantic. Silicate is less utilized by the local communities of diatoms, so it reaches the Lesser Antilles.

Seasonal MLD and nutrient concentrations covary over much of the data set, particularly in the northern regions. However, in terms of interannual variability, there is no robust relationship between MLD and nutrient anomalies in the data set in spite of vertical mixing being considered the main supply of nutrients to the surface ocean in the midlatitude ocean (Henson et al., 2003). Indeed, in some years such as 2008 and 2012 when the MLD is on average shallow relative to other years in the data set, the nitrate concentrations are also lower. However, counterexamples are the years of 2004 and 2011 when this relationship between MLD and surface nitrate does not hold (Figure 3d). Hence, there are additional factors that need to be considered. First of all, the variability of the ocean chemistry is influenced by physical as well as biological variability, and the upper ocean cannot be considered to be in a steady state (Glover & Brewer, 1988). Furthermore, in the lower latitudes of the NASTG, the dominant nutrient sources are horizontal advection from neighboring nutrient-rich regions, Ekman flux across the gyre boundary, mesoscale eddies, or other sources such as nitrogen fixation (Williams & Follows, 1998). A further issue has been identified by Palter et al. (2005) in that cold winters that promote deep convective mixing can actually diminish downstream primary productivity by enhancing the formation of the nutrient-depleted subtropical mode water that forms at the northern edge of the NASTG. Hence, although MLD remains a good indicator of the amount of convective mixing happening in the midlatitude Atlantic, as evidenced by the covariability with nutrient concentrations over seasonal time scales, it appears the interannual variability in nutrients could not be associated with MLD anomalies in a straightforward manner.

Whatever the source of nutrients, it is clear that they are consumed during the productive season. The nitrate concentrations drop to levels close to the detection limit every summer even in the northernmost region which shows the importance of biological uptake in determining nutrient budgets. We therefore analyzed satellite-derived chlorophyll-*a* and primary productivity. Using satellites to determine rates of primary production is the only available method of obtaining repeated global-scale measurements (Smyth et al., 2005). There are known issues such as the imperfect agreement between different satellites, the difficulty in distinguishing between trends, and noise for some biomes (Beaulieu et al., 2013) or underestimation since they only image the surface, whereas the maximum chlorophyll levels are usually seen at depth (Estrada et al., 2016). However, a recent study by Brewin et al. (2016) found a good match between satellite chlorophyll

estimations and in situ data along the Atlantic Meridional Transect cruise and concluded that the performance of satellite algorithms is actually better than described in previous studies. We therefore examined the satellite products over the same time and space intervals as our nutrient time series (Figure 3e).

Two years with higher average chlorophyll-*a* values between 30° and 35° N are 2010 and 2011. In fact, it seems the 0.25 mg m⁻³ contour for chlorophyll-*a* and the 1 μmol L⁻¹ contour for nitrate behave similarly. The differences between nitrate and chlorophyll are due to the different timings in the beginning of the productive season. While in the southern regions, the decrease in nutrient values begins early in the year, the nitrate values are still high in the northern regions, where chlorophyll-*a* values also remain high until later in the year. The NASTG is characterized by low chlorophyll-*a* concentrations throughout the year. The monthly mean chlorophyll-*a* concentration between 20° and 25° N only exceeds the 0.07 mg m⁻³ threshold that defines oligotrophy in 17 months throughout the entire time series (McClain et al., 2002). Two unusual summer blooms are identified in the southernmost latitudinal band during the summers of 2007 and 2010. This phenomenon has been studied before, and potential explanations include nitrogen fixation as a driver or vertically migrating phytoplankton (Wilson & Qiu, 2008).

3.2. Interannual Variability

Since the nutrient concentrations have a strong seasonality, especially at higher latitudes, they have a high degree of autocorrelation. The autocorrelation functions for the monthly nutrient concentrations displayed peaks at multiples of 12 month lags. We therefore expected the removal of seasonality to alleviate the issue of investigating long-term trends. Similarly to the nitrate and phosphate annual cycles presented, we calculated the mean annual cycles for the other variables used in this study. These were removed from the monthly averages, and Durbin-Watson tests were performed on both the averages and the anomalies. This statistic ranges in value from 0 to 4. A value near 2 indicates nonautocorrelation; a value toward 0 indicates positive autocorrelation; a value toward 4 indicates negative autocorrelation. The nutrients seasonal cycle in Region 1 has a small amplitude, so removing it did not alter the autocorrelation test value, which was already between 1 and 2. However, for the nutrient monthly averages in Regions 3–5, which had Durbin-Watson values smaller than 1, removing the seasonal cycle improved the results to values above 1. For the remainder of the study, we used nutrient anomalies in our discussion.

The nutrient anomalies are shown in Figure 4. Based on the average annual cycle of nitrate, the anomalies were grouped into “bloom,” “post-bloom,” and “pre-bloom” months and color coded (blue, black, and red, respectively). The bloom months were March–July, when nitrate usually decreased compared to the previous month. Nitrate concentrations near the limit of detection were recorded in the post-bloom months and increasing concentrations in the pre-bloom months. The intra-annual variability is too small in Region 1, so all values were plotted in black symbols. In addition, a comparison was made between SOO-derived nitrate anomalies and BATS nitrate anomalies, available online (<http://bats.bios.edu/bats-data/>). The latter were calculated by averaging the concentrations of samples collected within the mixed layer at the BATS station and subtracting the 2002–2016 monthly average. A similar pattern of elevated concentrations is seen toward the second part of the time series. However, they slightly precede the signal seen in the SOO data. More specifically, during the early part of the time series (2003–2006), there are a few elevated concentrations at BATS, up to 0.4 μmol L⁻¹ above the long-term average, slightly higher than the anomaly seen over the same time period in the record presented here. Much higher anomalies are seen at BATS during the latter part of the time series, mainly in 2010–2013. These are similar in scale to the significant increases seen in the SOO record presented here, however precedes them in time by a couple of years. This suggests that similar changes to those we observe occurred across broader areas of the subtropical gyre, although not at the same time. It is possible that they are linked to the postulated reductions in the AMOC strength documented elsewhere and that these begin on the western side before propagating to the interior where we observed them. Additionally, other data sets in Region 1 were examined, such as the annual Bermuda-Puerto Rico validation cruise (<http://batsftp.bios.edu/BATS/bottle/bvalbottle.txt>) or hydrographic cruises on the 24° N section (<https://cchdo.ucsd.edu/>). These were consistent with the observed trend; however, the lack of monthly sampling means that we cannot produce monthly anomaly plots similar to Figure 4.

Between May 2011 and February 2014, all the nitrate anomalies in Region 1 are positive, while a similar behavior is not observed in the phosphate anomalies. The explanations could be that (1) the spatial coverage of the sampling has changed, (2) phosphate is consumed at a higher rate than nitrate, (3) there are external

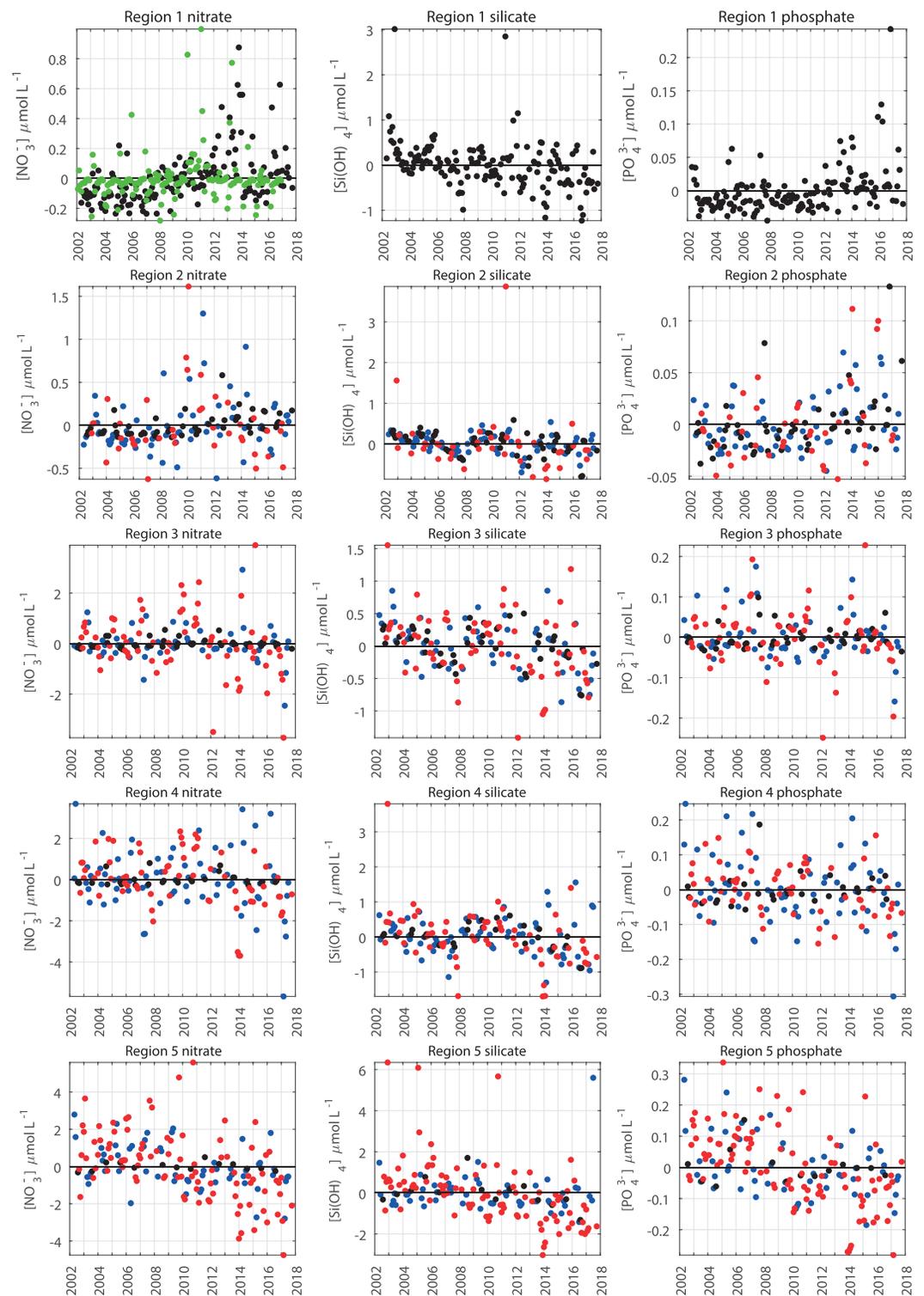


Figure 4. Monthly nutrient anomalies from the seasonally detrended data. The horizontal lines show the 0 value. The horizontal axes ticks and grid lines represent January of the respective year. The data points in Regions 2–5 are color coded according to the annual cycle: blue for bloom months, black for post-bloom months, and red for pre-bloom months. The nitrate anomalies at BATS (green symbols) are plotted together with the SOO nitrate anomalies in Region 1.

sources of nitrate which do not contain a proportional amount of phosphate, or (4) there is an input of a water mass containing more nitrate than phosphate.

It is unlikely the first explanation is the cause since the ship tracks have remained relatively stable with time. Indeed, excess phosphate is known to exhibit zonal gradients in the NASTG (Palter et al., 2011). However, the sampling strategy has remained constant with time, with samples collected in the southeastern limb of Region 1 during the outward journey and in the northwestern limb during the return journey less than 2 weeks later. As a further test in order to ensure the temporal changes are not artifacts of spatial variability, we also averaged the nutrient concentrations in a $2^\circ \times 2^\circ$ grid and subtracted these local means from the individual measurements to obtain a local anomaly. The same nutrient variability observed in the plots produced with region-wide anomalies was observed. The signal is therefore not a result of changes in sampling protocol.

It is also unlikely the second explanation is real since the surface NASTG is recognized as a nitrogen-depleted environment, hence the existence of a niche for N_2 fixers (Reynolds et al., 2007). Species of phytoplankton that are favored by low N:P ratios due to their different cellular content have been identified as “bloomers” (Arrigo, 2005). These organisms are adapted to exponential growth and are usually found at higher latitudes. In environments with scarce resources such as Region 1, the optimal N:P cellular ratio of the “survivalists” is >30 , and the uptake ratio of N:P has also been found to be higher than 16:1 (Martiny et al., 2013). Moreover, phytoplankton biomass and productivity in the NASTG usually increase following the addition of nitrate during in situ and mesocosm experiments (Moore et al., 2013). It is therefore unlikely the reason why nitrate accumulates is the lack of consumption by surface organisms which would involve a shift in phytoplankton community structure.

The processes that could contribute to the third explanation are atmospheric nitrogen deposition and nitrogen fixation. Atmospheric nitrogen deposition, which is predominantly anthropogenic in origin, is thought to account for up to 3% of the annual global marine new biological production and up to 20% in upwelling-limited regions such as the NASTG (Duce et al., 2008). These effects are expected to continue to grow in the future. However, feedback mechanisms in the nitrogen cycle would be expected to cause nitrogen fixation to be negatively affected, so the associated increase in marine productivity may be suppressed (Fernandez-Castro et al., 2016; Somes et al., 2016). Furthermore, atmospheric deposition only significantly influences surface nitrogen concentrations during the summer (St-Laurent et al., 2017).

Nitrogen fixation is a mechanism specific to diazotrophic organisms, especially *Trichodesmium* sp. Nitrogen fixation has traditionally been assumed to require warm, well-stratified and iron replete waters to thrive (Fernandez-Castro et al., 2016), but that view was recently challenged by studies that found high N_2 fixation rates even in poorly stratified waters (Tang et al., 2019). The deficiency of fixed nitrogen relative to phosphorus is not a sufficient condition for nitrogen fixation (Codispoti et al., 2001). Nitrogen fixation occurs in the central part of the NASTG as suggested by the low isotopic signature of the nitrogen in the organic matter produced in this area (Mompean et al., 2016). This location is suitable due to the proximity of the Saharan desert and in particular the Bodélé Depression as a supply of iron (Moskowitz et al., 2016). Whether this process is the reason behind the observed increasing trend in nitrate in Region 1 of our data set is difficult to say. The amount of Saharan dust drawn depends on the strength of atmospheric pressure systems over the Atlantic (Goudie & Middleton, 2001), but increases in production associated with nitrogen fixation have only been identified in the summer blooms (Wilson & Qiu, 2008). The surface nitrate concentration and therefore primary production is mostly enhanced indirectly by the subsequent remineralization of the fixed nitrogen added to the system by diazotrophs. Also, N_2 fixers require phosphorus for their metabolism, so the lack of phosphate in the surface water could become a limiting factor.

As for the final explanation, a change in water mass distribution and/or circulation would be a likely mechanism. The thermocline of the North Atlantic is unique in having a positive N^* (Fanning, 1992). Processes such as nitrogen fixation or atmospheric nitrogen deposition (discussed above) that occur in the NASTG lead to sinking organic matter enriched in nitrogen. Moreover, dissolved organic phosphorus is more bioavailable and sometimes directly utilized by primary producers at the surface (Letscher et al., 2015; Mather et al., 2008). Consequently, the dissolved organic matter is becoming enriched in nitrogen relative to phosphorus with depth (Abell et al., 2000). If this water mass gets brought to the surface through changes in circulation patterns, the excess supply of nitrate could be explained. Deeper mixing is unlikely to be the reason for the observed positive nitrate anomalies. The winter of 2011/2012 was a season of weak mode water formation in

the NASTG, which led to shallower than usual MLD throughout the gyre and potentially less nitrate injected (Billheimer & Talley, 2013). However, the nitrate anomalies in 2012 are all positive. Alternatively, since the Gulf Stream has been identified as a nutrient stream (Williams et al., 2011), changes in the transport of nutrients out of the subtropics related to AMOC variability could impact subtropical nitrate budgets.

There is substantial interannual variability in the data set. In the more recent years of the time series, phosphate levels have increased at a rate higher than the Redfield ratio when comparing it with the nitrate values. Therefore, the N^* values for the surface ocean are mostly negative. The extra phosphate in the surface may enhance nitrogen fixation rates which could explain the rising N:P ratio of the sinking flux observed at 150 m depth at BATS (Lomas, 2018).

Variability exists in the silicate anomalies as well, but it appears concentrations have been dropping. Regionally, changes are likely dominated by variability in convective mixing. A similar decreasing trend in silicate concentrations was observed in the subpolar North Atlantic (Hátún et al., 2017). This change was attributed to decreased winter convection, a weaker subpolar gyre, and a higher influx of subtropical origin water. Although surface silicate concentrations have gone down, there was no observed effect on diatom abundance. Continuous Plankton Recorder (CPR) total diatom abundance data from the Sir Alister Hardy Foundation for Ocean Science (Johns, 2018) were averaged spatially and temporally, and measurements were normalized by the sampling effort. Since the plankton tows usually happen at midlatitudes, there were not enough samples taken in Region 1 of this study. There is no statistically significant trend observed in any of Regions 2 to 5 between 2002 and 2016 although strong interannual variability is observed. However, CPR data should not be used as an absolute measure of abundance but as a semiquantitative estimates (Richardson et al., 2006). In addition, averaging CPR data over large areas can introduce biases. Nevertheless, we found no conclusive evidence for a relationship between the surface silicate concentrations and diatom abundance.

3.3. NPP

Region 1 is observed as being nitrate deficient throughout the year (Figure 2), but the recent positive periods of apparent positive nitrate anomalies from 2011 to 2014 should have caused an alleviation of this limitation and led to increased net community production. We might expect any associated increase in net community production to be accompanied by a correspondingly higher phytoplankton biomass and increased net primary production. We used data produced by a Vertically Generalized Production Model using surface satellite chlorophyll-*a* concentrations, sea surface temperature, and photosynthetically active radiation to test this (Behrenfeld & Falkowski, 1997), and we resorted to using NPP to look for potential effects of the nutrient variability.

Contrary to what might be expected on the basis of the increasing nitrate concentrations, the NPP does not increase in the 2011–2013 part of our time series, when nitrate anomalies were positive. Instead, all the annual average values within our time series ranged between 190 and 212 $\text{mg C}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$ apart from the Years 2010 and 2011 when they reached 240 and 219 $\text{mg C}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$, respectively. A possible explanation for the NPP not responding to increases in nitrate is the reduced importance of the inorganic form of the nutrients at these low latitudes compared to the organic one. It seems that even though nitrate is increasing, it does not have a long-term effect on primary production since about 40% of the particulate nitrogen exported over the gyre is supported by semilabile DON (Torres-Valdés et al., 2009). In addition, the f ratios at these latitudes in the Atlantic are around 0.2, so even a large change in new production might only result in a small change in net primary production (Planas et al., 1999).

In order to test the nutrient limitation regime of the NASTG, a shipboard incubation experiment was performed in Region 1 during the fall season of 2017. The initial chlorophyll concentrations were found to be between 0.04 and 0.06 mg m^{-3} , values typical for the surface of an oligotrophic area of the ocean (Figure 5). In the control bottles, with no nutrient addition, the mean chlorophyll concentration decreased in all three experiments (one of them significant), while the mean phaeopigment concentration increased in two out of the three experiments (none significant). The phaeopigment:chlorophyll ratio has the highest mean value in the control bottles in all three experiments. This indicates that in the 48 hr of the experiment, the phytoplankton exhausted the available nutrients and that due to a combination of nutrient stress and potential grazing, degradation products began forming. Addition of nitrate caused an increase in chlorophyll in all three experiments, but only in Experiment 2 is the mean significantly different from the control (tested

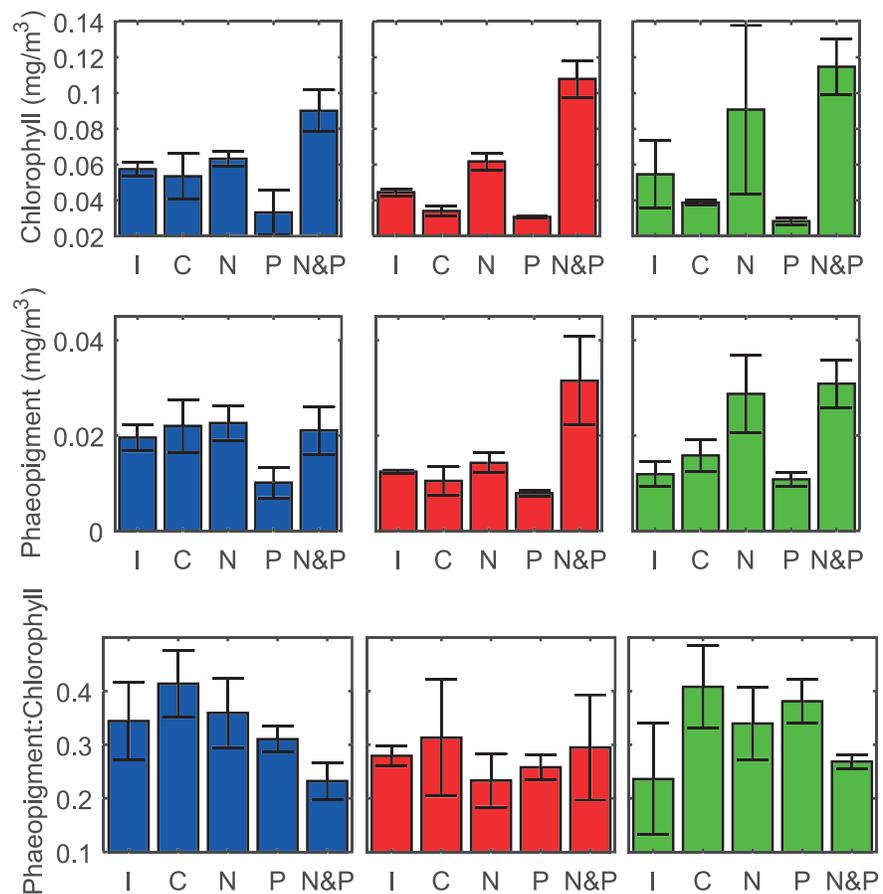


Figure 5. The results (\pm standard deviation) of the nutrient limitation experiment showing chlorophyll-*a* concentrations (top row), phaeopigment concentrations (middle row), and the ratio of the two (bottom row). The experiment was performed three times at different locations. For each experiment, water was filtered for analysis at the start of the experiment (I) and 48 hr later with no nutrient addition (C), nitrate only addition (N), phosphate only addition (P), and nitrate + phosphate addition (N&P). Each column represents a different location where water for the experiment was collected. Blue, red, and green colors represent Experiments 1, 2 and 3, respectively.

using a one-way analysis of variance and a post hoc Tukey test, where *p* values lower than 0.05 were considered significant). Addition of both nitrate and phosphate caused an even larger increase in chlorophyll, statistically different from the control bottles in all three experiments (analysis of variance). This reinforces past knowledge that the surface NASTG is primarily limited by nitrogen and secondarily by phosphorus (Davey et al., 2008; Mills et al., 2004; Moore et al., 2008, 2009). Surprisingly, addition of phosphate alone caused a decrease in chlorophyll, albeit not significant. However, it was not accompanied by an increase in the phaeopigment:chlorophyll ratio. This sort of behavior has been observed before in an experiment in the Mediterranean Sea. The explanation proposed is a “trophic bypass,” whereby the added phosphorus bypasses the phytoplankton through a predatory food chain from heterotrophic bacteria (Thingstad et al., 2005). This produces an increase in small-celled phytoplankton loss through predation. While these results show that during the fall of 2017, the NASTG was colimited by nitrate and phosphate; they do not dismiss the possibility that at times, such as the 2011–2014 period when all the monthly averaged nitrate anomalies are positive, the system could shift to a different regime.

3.4. Interannual Variability in NPP

Substantial interannual variability in NPP is prevalent across the North Atlantic (Figure 6), with high values in NPP recorded in 2010 and 2011 in both Regions 1 and 2 (and corresponding with the more southward extension of the chlorophyll-*a* contours in the Hovmöller diagram in those 2 years as shown in section 3.1). The shape of the anomaly plots are similar even if only the spring productive months are considered rather than the annual production, suggesting that changes in the spring production are the main contributor to

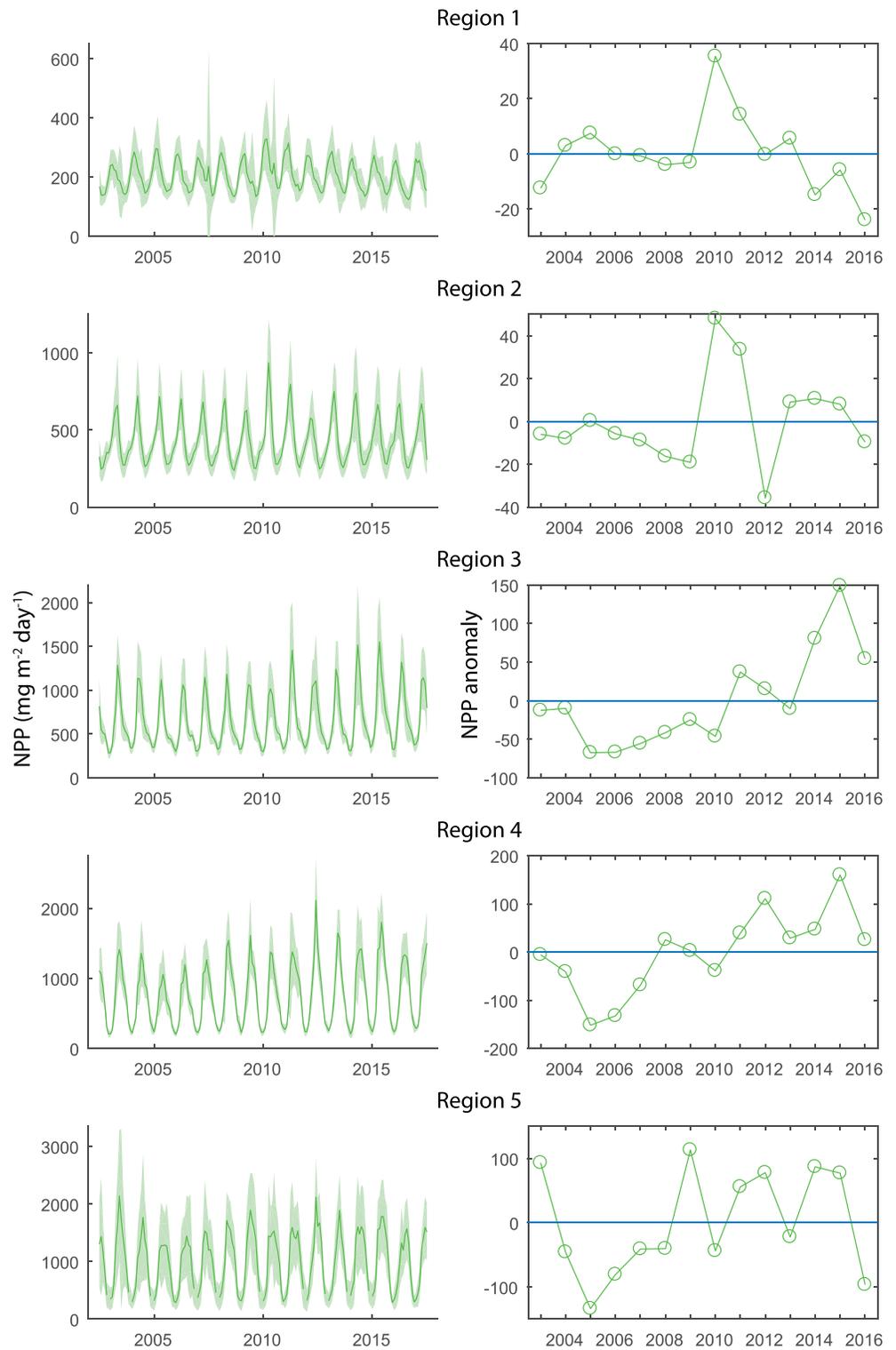


Figure 6. Monthly averages (\pm standard deviation as shading) of satellite-derived net primary productivity expressed as $\text{mg C}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$ (left) and the annual anomalies (right) for the five regions of this study.

Table 2

New Production (P_N) and Net Primary Production ($\int NPP$, Yearly Integration of Net Primary Productivity) Expressed as $g\ C\ m^{-2}$ for the Years of 2010 and 2011 as Well as the Mean \pm Standard Deviation for the 2003–2016 Period (Mean) and the Mean \pm Standard Deviation for the Same Period With the Years of 2010 and 2011 Excluded (Mean**)

	Region 1		Region 2		Region 3		Region 4		Region 5	
	P_N	$\int NPP$	P_N	$\int NPP$	P_N	$\int NPP$	P_N	$\int NPP$	P_N	$\int NPP$
2010	—	87.7	20.5	175.0	60.9	220.3	75.2	260.4	66.8	347.5
2011	—	80.0	10.5	169.8	39.7	250.7	63.7	288.8	33.3	383.9
Mean	—	74.8 \pm 5.1	5.9 \pm 4.9	157.5 \pm 7.8	32.3 \pm 12.1	237.2 \pm 22.7	57.4 \pm 15.5	274.5 \pm 30.7	63.1 \pm 30.0	363.5 \pm 29.6
Mean**	—	73.3 \pm 3.3	4.3 \pm 2.1	155.0 \pm 4.8	29.3 \pm 9.1	237.5 \pm 23.7	55.4 \pm 15.7	274.5 \pm 32.9	65.2 \pm 31.3	363.1 \pm 31.3

annual variability. It is likely anomalously high concentrations of sea surface nitrate drive this variability since the peaks in the chlorophyll-*a* plot happen in the spring growth seasons immediately after the nitrate is brought to the surface in the winter mixing season. The nitrate anomaly only appears as distinct peaks at latitudes higher than 30° N. This is why the nitrate anomaly plot for Region 1 (latitudes lower than 31° N) does not display the double peak in the Years 2010 and 2011.

We calculate how the extra supply of nitrate is contributing to new production in the two anomalous years in four of the regions as follows. New production can be estimated when only sea surface nutrient concentrations and MLDs are available (Frigstad et al., 2015; Henson et al., 2003; Wong et al., 2002). First, we estimate annual nitrate consumption as

$$\Delta N = [N_{\text{spring}} - N_{\text{summer}}] \times [(MLD_{\text{spring}} + MLD_{\text{summer}})/2], \quad (4)$$

where N_{spring} and N_{summer} are the concentrations of nitrate at the start and end of the productive season, respectively—the maximum and minimum nitrate concentrations and MLD_{spring} and MLD_{summer} are the MLDs for the respective months at the start and end of the productive season. New production is then

$$P_N = R\Delta N, \quad (5)$$

where R is the Redfield ratio of carbon to nitrogen in phytoplankton. In this study we used a value of 6.6 for the R factor. Integrating the nitrate concentration difference over the mean MLD for the productive season provides a first-order approximation of the nitrate consumed. The estimated new production (with units converted from moles to grams) and the net primary production as a context are shown in Table 2.

In Region 2, the new production was on average $4.3 \pm 0.6\ g\ C\ m^{-2}$ for the duration of our time series excluding the Years 2010 and 2011. New production was 20.5 and $10.5\ g\ C\ m^{-2}$ in 2010 and 2011, respectively. This extra new production seems to match most of the increase seen in satellite-estimated net primary production (calculated by integrating the NPP over each year). The net primary production was $175\ g\ C\ m^{-2}$ in 2010 and $170\ g\ C\ m^{-2}$ in 2011 compared to the long-term mean value of the other years of $155.0 \pm 4.8\ g\ C\ m^{-2}$. Thus, the scale of the increases in the nitrate-based estimate of new production and the chlorophyll-based estimate of net primary production are different with a twofold to fourfold versus a 13% increase, respectively. This reflects the fact that new production is much smaller than net primary production, by a factor of around 5 in this area of the ocean.

In Region 3, new production was higher in 2010 ($60.9\ g\ C\ m^{-2}$) and 2011 ($39.7\ g\ C\ m^{-2}$) than the average value for the other years ($29.3 \pm 2.6\ g\ C\ m^{-2}$). However, the net primary production for these two years (220 and $250\ g\ C\ m^{-2}$) was within the range of variability of the long-term mean value of the other years ($237.5 \pm 23.8\ g\ C\ m^{-2}$).

In Region 4, new production was also higher in 2010 ($75.2\ g\ C\ m^{-2}$) and 2011 ($63.7\ g\ C\ m^{-2}$) than the average value for the other years ($55.4 \pm 4.5\ g\ C\ m^{-2}$). Since the Porcupine Abyssal Plain Observatory is situated at 49° N and 16.5° W within our defined Region 4, we can compare our values with other studies performed at this location. Our values are higher than the new production obtained from nitrate consumption by Frigstad et al. (2015) of $29.3\ g\ C\ m^{-2}$ (we converted their $0.37\ mol\ N\ m^{-2}$ to $g\ C\ m^{-2}$) but comparable to the value for net community production of $54.8\ g\ C\ m^{-2}$ they obtained using dissolved inorganic carbon drawdown. The 2010–2011 events thus increased new production in Regions 2–4 of our study area. The smaller variability in net primary production might be expected as this will be the sum of both the production fueled by any new

introduction of nutrients to the surface layer and all of the productivity supported by nutrient regeneration within this layer.

New production in Region 1 is not calculated in Table 2 since, according to equation (4), the difference between peak winter and minimum summer nitrate is used. In Region 1, the seasonality is very low (Figure 2), and a clear period of new nitrate utilization is difficult to discern. In Regions 2–4, however, the new production becomes a larger proportion of the net primary production in 2010 and 2011 compared to the long-term ratio, suggesting that nutrients brought to the surface by such events are rapidly utilized, such as the observations directly indicate.

We now investigate the possible causes for the observed anomalies in 2010 and 2011. The nutrients are brought to the surface during the winter season, so we checked whether enhanced winter mixing through a deeper MLD could be the cause of the anomalous NPP. Weak but statistically significant Spearman's rank correlations are observed between nitrate and MLD anomalies in Regions 3 and 4, with correlation coefficients of 0.20 and 0.17, respectively. However, the deepening of the MLD is only seen in 2010, whereas the nitrate anomaly persists during the winter of 2011 as well. In addition, the maximum MLD was only 10 to 35m deeper in the winter of 2010 compared to other winters, so it is unlikely this is the sole process responsible for the nutrient anomaly at the surface. MLD was found to be insufficient as a predictor of productivity in the North Atlantic subtropics (Lozier et al., 2011). The depth of the nitracline could alternatively be important.

Variations in the NAO, a characteristic of the atmospheric circulation over the North Atlantic, could impact the formation of waters that are ultimately the source of nutrients in the subtropical thermocline. A positive index is usually related to increased intensity of westerly winds across the midlatitude North Atlantic (Delworth et al., 2016). Historically, over the last century, the NAO has been in a mostly positive state, but more recently, it has returned to neutral values on average. Only in the last decades has the sea surface temperature of the North Atlantic been significantly correlated with the NAO index (Walter & Graf, 2002). This long-term change may have induced significant regional changes in the upper ocean nutrient supply (Oschlies, 2001). Lower values of the NAO are associated with deeper mixed layers and larger entrainments of nutrients into the surface ocean at BATS and across the western NASTG (Gruber et al., 2002; Patara et al., 2011). Counterintuitively, a negative NAO can also mean a decrease in nutrients downstream of the formation of a thicker nutrient-depleted subtropical mode water (Palter et al., 2005). Lomas et al. (2010) found however this effect to be of minor importance in explaining the observed patterns. They attribute the increase in nutrients to a more continuous but not necessarily deeper mixing in NAO years in the NASTG. The NAO's effect on nutrients and therefore on biological production has been observed at higher latitudes as well, as far as the subpolar gyre (Henson et al., 2013). Indeed, the anomalous years in our data set coincide with strong negative NAO index years.

Lastly, variability in circulation patterns may impact the advected distribution of nutrients at the surface. The AMOC is a key driver of the circulation patterns in the North Atlantic and is the source of the transport of nutrients from the Southern Ocean that fuels biological activity in the region (Williams et al., 2000). In 2009–2010, a 30% decrease in northward transport of heat at 26° N was identified as the cause of a coldwater anomaly in 2010 that then reemerged in 2011 (Bryden et al., 2014; Cunningham et al., 2013). This was attributed to a decrease in the southward transport of deep waters (Smeed et al., 2014) and the beginning of a shift to a lower state of circulation strength observed over the last decade (Smeed et al., 2018). A drop in northward heat transport and its impact on the position of the Gulf Stream (as highlighted earlier, a key source of nutrients for biological activity in the NASTG) could well have impacted the accumulation and distribution of nutrients in surface waters.

4. Summary

We present a high temporal and spatial resolution data set of sea surface inorganic macronutrient measurements in the subtropical and midlatitude North Atlantic between 2002 and 2017. The use of regular sampling onboard commercial ships trading along repeat routes allows monthly monitoring and the development of time series where seasonal through interannual variability can be studied across large ocean regions. Although comprehensive, we acknowledge that the length of this time series is nowhere near long enough to match the ≈ 30 years globally averaged minimum length of time estimated to be needed to observe surface nitrate in order to detect a climate change-driven trend above background variability (Henson et al., 2016).

However, our 15 year time series has already produced important results and will continue to grow in the future since the project is still ongoing.

On average, we observed surface nitrate depletion at the end of the productive season with both nitrate and phosphate increasing northward in our study area. The seasonality is also more pronounced in the northern regions. We observed significant interannual variability in the data, non-Redfieldian long-term increases in nitrate and phosphate in the subtropical gyre, and decreases in silicate throughout the study area. We presented the most recent evidence of the nutrient limitation regime of the NASTG through a shipboard incubation experiment. We observed an anomalous event in the winter seasons of 2010 and 2011 with nitrate concentrations higher by more than $1\mu\text{mol L}^{-1}$ compared to the long-term mean that led to increased values of new production across our study area.

Acknowledgments

We thank the two anonymous reviewers whose comments/suggestions helped improve and clarify this manuscript. We would like to thank the following individuals and organizations: the captains, officers, and crews of MV *Santa Lucia*, MV *Santa Maria*, and MV *Benguela Stream* for continuous technical assistance and support onboard the ships, staff of Geest Line, U.K., and Seatrade Reefer Chartering, Belgium, for supporting the project; the scientific and technical crew part of the BATS team on board the BVAL 52 cruise for their help with the nutrient limitation experiment; all the analysts and lab technicians that analyzed the samples throughout the years; IFREMER/LOS Mixed Layer Depth Climatology website (www.ifremer.fr/cerweb/deboyer/mld) and UCSD Mixed Layer Depth website (<http://mixedlayer.ucsd.edu/>) for access to mixed layer depth data; NASA Ocean Biology Processing Group (OBPG) (<https://podaac.jpl.nasa.gov/>) for access to MODIS satellite-derived chlorophyll concentrations; BATS (<http://bats.bios.edu/bats-data/>) for access to their nitrate data set; and Ocean Productivity website (<http://www.science.oregonstate.edu/ocean.productivity/>) for access to NPP data. The nutrient data are publicly available from the British Oceanographic Data Centre (https://www.bodc.ac.uk/data/published_data_library/catalogue/) with a DOI of doi:10.5285/7a7b497a-c47b-0a69-e053-6c86abc07ef0. Recent work on the line has been supported by the NERC Greenhouse Gas TAPNE/k00249x/1 project. The work on this paper was supported by a studentship from the Natural Environmental Research Council (Grant NE/L002531/1).

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