# Industrial-era decline of subarctic Atlantic productivity

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## Supplementary Information

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# I. Supplementary Sections

## S1. Marine airmass source attribution and covariation in Greenland [MSA] records

### *a. Marine airmass source attribution by site*

A notable degree of low-frequency covariability is observed amongst preexisting methanesulfonic acid-concentration ([MSA]) records from Greenland (Extended Data Fig. 4). Prior authors have independently identified a c. 150-200 year decline in [MSA] in the 19$^{th}$-20$^{th}$ centuries across several ice core [MSA] records[18,31,34], though its attribution has thus far remained uncertain. Due to i) the particularly short atmospheric residence time of MSA (~7 days, a function of the species' high hygroscopicity and susceptibility to particle scavenging[62]), and ii) known differences in the primary moisture source of airmasses arriving to Greenland[63,64], broad covariation of [MSA] across Greenland is not necessarily expected[65].

In order to better understand the physical basis behind the observed [MSA] covariation, we analyzed summertime (JJA) trajectory patterns of atmospheric air parcels arriving at each ice core site in our Greenland array using the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT), version 4.9 (ref. 50). We assumed the primary source region of Greenlandic-deposited MSA to be commensurate with the most-probable summertime (JJA) trajectory path(s) taken by low-lying, oceanic (i.e., existing within the marine boundary layer) air parcels traveling en route to each ice core site. Due to positioning errors of individual trajectories, which increase with distance from the particle receptor sites[46], we adopted a probabilistic approach here, in which a large-number of 7-day back-trajectories were computed (one particle released each day at 12:00:00 PM from AD 1948-2013) for each site and integrated into probabilistic marine airmass transport density maps. These maps, normalized on a 0-1 relative scale, provide an index of the relative probability (0-1 = least to most probable, respectively) that a given atmospheric particle would travel over a given oceanic grid box at a given time. Using information of atmospheric height (above sea level) recorded for each particle, we similarly computed mean trajectory elevations for each latitude-longitude bin.

The JJA marine-airmass transport density maps for each site can be viewed in Extended Data Figure 5. Qualitatively, the sites appear to lie along a spectrum, whereby sites situated more southerly in Greenland receive predominantly easterly- to southeasterly-sourced (the Irminger Sea and Subarctic Gyre (SPG) regions) trajectories, while sites situated further north on the GrIS exhibit a predominantly westerly-sourced trajectory pattern.

To more-objectively differentiate the sites by their varying airmass transport densities (Extended Data Figure 4), we performed factor analysis with varimax rotation[66] on each site's airmass transport density field under the *a priori* assumption of two underlying airmass source regions, or "factors". The two inferred factors, as qualitatively identified above, are assumed to represent either 1) the Irminger Sea/SPG or 2) Baffin Bay/Labrador Sea as primary airmass sources. The results of the experiment are summarized by analysis of the squared loadings, or communalities, shown in Fig. 1b, which represent the fraction of the variance explained by each factor (columns) at each site (rows). For 11/12 sites, over 90% of the variance is explained, indicating our *a priori* assumption of retaining two primary MSA source regions to be sufficient (the remaining site, 20D, has >80% of its variability explained). As expected, the factor analysis correctly identifies the most southerly GrIS sites (20D and GC; Extended Data Figure 4) as having the highest Factor #1 scores, while the most northerly GrIS sites (e.g., NGTB20, NGTB26, TUNU) score highest on the Factor #2 loading. Grouping the sites as either Factor #1 or #2 sites based on the highest communality score achieved by each site indicates the seven most-southerly GrIS sites – 20D, GC, D4, Summit2010, GRIP93a, NGRIP, and B16 – are primarily derived from the Irminger Sea/SPG, while airmasses arriving at the five most-northerly GrIS sites – NGTB18, NGTB20, NGTB21, NGTB26, and TUNU – may be of predominantly Baffin Bay/Labrador Sea origin (notwithstanding notable similarities in all sites' median atmospheric elevation grids; Extended Data Figure 5).

Importantly, analysis of the Factor #1 and Factor #2 communalities also indicate that airmass sources across GrIS sites are not wholly independent, but instead represent variable mixtures of easterly

(Factor 1) vs. westerly (Factor 2) sourced airmasses. As summarized by Figure 1b, a Factor #1 score of 1 roughly represents 100% of airmass trajectories sourced from the Irminger Sea, while a Factor #2 score of 1 roughly indicates 100% of airmass trajectories sourced from Baffin Bay/Labrador Sea. Since no sites displayed a communality score as high as 1, this – in combination with similarities found between the median particle elevation maps – suggests a common [MSA] signal should be embedded within all Greenlandic sites. In the following section, we explore this prediction more explicitly by analyzing the [MSA] records produced for each site.

*b. Exploratory principal component analysis of Greenlandic [MSA] records*

To explore the degree of similarity amongst the Factor #1 and Factor #2 sites' [MSA] records, we plot in Extended Data Figure 6 the unit-variance standardized and mean-centered [MSA] time series (relative to A.D. 1821– 1985) for all 12 sites over the time period A.D. 1767 – 2013, the oldest year still containing ≥75% (at least 9/12) of the original records. If composited into either Factor #1 or Factor #2 sites, remarkably high coherence, even at interannual-scales, is found in the two time series ($r^2$ = 0.40, $p <$ 0.0001; ref. 55). A notable feature amongst records from both airmass source regions is the conspicuous ~150-200 year decline in [MSA], beginning at c. A.D. 1800-1850, and occurring into the late 20$^{th}$ century.

To better extract mode(s) of covariability amongst individual records, we performed an exploratory principal component analysis (PCA; Section S2) using records from both airmass source regions. PCA is a commonly used Eigen decomposition technique, which allows one to linearly re-project a dataset as a new set of basis vectors, whereby each new projection represents a mutually-orthogonal representation of the former while representing a progressively smaller variance fraction of the original dataset (e.g., ref. 67). We again limited the analysis from the most-recently available data (i.e., A.D. 2013) until A.D. 1767. As some records have missing data at their extremities, a statistical data-infilling procedure was used for the PCA analysis[42], as elaborated in the Methods portion of the main text, and the next section (S2).

Results of the PCA for the Factor #1 and Factor #2 [MSA] records, revealed a slightly improved degree of similarity (not shown; $r^2$ = 0.43, $p$ < 0.0001; ref. 55) amongst each factor's first principal components (PC1), which capture ~50 and 33% of the variability, respectively. The significant correlation found between the two PC1's is also impervious to linear detrending (ref. 55), underscoring the high degree of interannual-decadal scale covariability captured by the Eigen decomposition. Conversely, no tailing modes of [MSA]-variability (including permutations therein) were significantly correlated, suggesting these modes represent locally confined bioproductivity signals, or "noise", amongst the records. The analysis provides robust evidence that PC1 successfully captures a common mode of Greenlandic-wide MSA variability, as independently predicted by the joint distributions of each Factor's airmass transport probability densities. As such, using the combined Greenlandic PC1 signal (capturing a median/mode of ~44% of the variability amongst the 12 records) we consider the Greenlandic sites as a single "MSA receptor" (Fig. 2).

*c. Spatiotemporal stability of marine-airmass trajectories and Arctic-influence*

While recent reconstructions have suggested more frequent and intense intrusions of upper-atmosphere Arctic "jet stream" meanders into the mid-latitudes since the mid 1960's (e.g., ref. 68), current evidence does not suggest such changes have had a discernable effect on the background state of subarctic Atlantic-derived bioaerosol deposition over the GrIS during our ~250-year study period. We explored this argument explicitly over the period AD 1948 to 2013 by systematically cataloguing the annual percentage of hourly oceanic trajectory-endpoints situated at latitudes further north than each ice core site's location. We found no sites showing long-term trends in northern-derived trajectories significantly different than zero ($p$ < 0.05), nor evidence of shifting intensities in Arctic-derived airmasses, despite the existence of multidecadal trends in GrIS-[MSA] over the same time period. More broadly, our results underscore prior studies that have analyzed the source of precipitating Greenland airmasses in equable[69-70], as well as substantially varied, background climate states[71] that each show consistency in the North Atlantic as a stable source of marine aerosols and moisture to Greenland.

## S2. Comparison of data infilling methodologies for [MSA]-PC1 computation

Prior to computing the PCA of the 12 Greenland [MSA] records over the period A.D. 1767-2013, multiple records required data infilling at their extremities. The method for data infilling used is described at length in the Methods portion of the main text. Here, we compare two additional methods for data infilling in order to test the veracity of [MSA]-PC1: 1) a Probabilistic PCA ("PPCA") algorithm is tested, which relies on the expectation maximization (EM) algorithm[72], as well as 2) a nested PCA-compositing approach, a hybrid methodology incorporating the EOF-based imputation algorithm[42] discussed in the main text.

We briefly describe the steps taken in applying the two methodologies for computation of [MSA]-PC1 and its uncertainty:

*PPCA*: Details on the PPCA algorithm (including its derivation and technical considerations for implementation) are described at length in multiple previous studies[73]. In general, PPCA can be viewed as a probabilistic extension of classical PCA, one that iteratively seeks the principal $q$ axes of an observed $d$-dimensional dataset (where $q < d$) that maximizes variance within a projected subspace, by explicitly incorporating maximum likelihood estimates of an isotropic (Gaussian) error model[74]. We take advantage of the MATLAB™ function "ppca.m" (available since R2013a in the Statistics and Machine Learning Toolbox) for implementation of the PPCA algorithm. As described in the Methods, we use a PCA-bootstrap routine ($n$ = 10,000 realizations of [MSA]-PC1 based on random sampling with replacement of the 12 [MSA] records[43]) to estimate confidence limits from the PPCA-derived [MSA]-PC1 signal.

*Nested EOF*: The nested PCA approach combines classical PCA, and the EOF data infilling routine. Specifically, we take an approach identical to that described in the main text for the imputation of missing values[42], of missing values since A.D. 1821, the oldest year represented by all 12 records (as such, the infilled data represents the leading modes of variance for the period A.D. 1821-2013). Following infilling of missing values (for which $m$ = 1,000 realizations were conducted, see Methods), we conducted PCA analysis within a nested loop, in which for each nest a PCA was conducted using a successively smaller number of records, based on temporal availability of the records. Following computation of the principal components in each nest, each PC1 was checked for qualitative and statistical consistency during periods of common overlap with the former nest's PC1. Provided consistency, the PC1 components were then spliced together. In total, our criterion of >75% record retention for computation of [MSA]-PC1 required only 4 nests to be computed. We again use the PCA-bootstrap routine (random sampling with replacement of $n$ [MSA] records, where $n$ = 12, 11, 10, and 9 for successive nests[43]) for confidence level estimation.

All three methods for PCA with missing data are essentially identical ($r > 0.99$), although PPCA generally tends to produce slightly more conservative uncertainty estimates (not shown). While we ultimately chose to use the EOF-based infilling routine[42] for its slightly more transparent methodology and interpretation (relying, at its core, only on fundamental concepts of linear algebra), conclusions of the study do not appear overly sensitive to which PCA-method is used in calculating [MSA]-PC1.

## S3. Satellite-derived net primary productivity (NPP) and temporal trends (1998 – 2016)

In this section, we provide a brief overview, including a model- and satellite-based inter-comparison, of NPP trends in the subarctic Atlantic. NPP is the rate of photosynthetic derived carbon fixation minus the rate of (bioavailable) carbon respiration by autotrophic communities. It has been estimated using satellite observations (~8 day reoccurring) for nearly ~2 decades, though is by itself not a property that can be measured directly from satellite platforms. Rather, estimates of NPP are based on empirically derived algorithms incorporating satellite-retrieved estimates of ocean physical properties such as (albeit not necessarily including or limited to) temperature, color (reflected wavelength), and (or) incident (photosynthetically-active) radiation.

We compared NPP using output from two independent algorithms: i) the (standard) Vertically Generalized Production Model (VGPM, ref. 12), and ii) the Carbon-based Production Model (CbPM; ref.

57). Both products include overlapping estimates of (monthly-contiguous) NPP estimates (SeaWiFS: Oct 1997 – Dec 2007; MODIS-Aqua: Jul 2002 – Dec 2017), and are available at mean-monthly 1/6˚x1/6˚ gridded resolution. Models i) and ii), and variants therein, are among the most commonly used models of NPP[75-76]. The VGPM model is primarily chlorophyll- $\alpha$ based and temperature dependent (parameterization of photosynthetic efficiency with sea-surface temperature based on a polynomial relationship[12]; for a comparison of VGPM variants incorporating different parameterizations, see ref. 76). Model ii) represents a complimentary, chlorophyll-independent description of NPP. The CbPM relates satellite-retrieved particle back-scattering variations to planktonic biomass concentration, which is in turn related to planktonic growth rate (i.e., NPP) via chlorophyll-carbon (planktonic biomass) ratios[57]. More details on the intercomparison of the models, and additional references therein, can be found linked via the URL in Table S3, or in the supplementary of ref. 75.

We assessed trends in subarctic Atlantic (50-65˚N, 60-10˚W) NPP using both models (VGPM and CbPM) and satellite sensors (SeaWiFS and MODIS-Aqua). The results are encapsulated in Extended Data Fig. 1. All models and satellite sensors show peak subarctic Atlantic productivity occurring during the summer months, Jun-Aug; note that partial polar darkness occurs over the subarctic Atlantic during the months Nov-Dec-Jan-Feb, which results in underestimates of NPP during those months. Inconsistencies between MODIS and SeaWiFS-derived NPP estimates (temporal overlap c. 2003-2007) are greatest for the CbPM-based NPP estimates, which show substantially higher NPP yields in the MODIS-based estimates. Note however, that although discrepancies between SeaWiFS and MODIS are comparatively minor for the VGPM-based NPP estimates, SeaWiFS-based annual NPP yields are systematically ~2-7% higher than MODIS-derived yields during all years of overlap. In Extended Data Figure 1 we also show alternate realizations of the weighted-least squares regression analysis of Fig. 3b (see methods of main text), comparing the SeaWiFS-VGPM, MODIS-VGPM, SeaWiFS-CbPM and MODIS-CbPM against contemporaneous [$DMS_{SW}$] measurements. Our results show that VGPM-NPP estimates are more closely related to [$DMS_{SW}$] than CbPM-NPP estimates, and SeaWiFS-based NPP estimates are more closely related to [$DMS_{SW}$] than MODIS-based NPP estimates. All four regressions, however, show statistical significance at the $p < 0.001$ level.

Due to superior inter-satellite sensor comparisons and improved covariation with [$DMS_{SW}$] (Extended Data Fig. 1), we report NPP trends deriving from the standard VGPM product within the main text. Due to the strong spatiotemporal consistency in both SeaWiFS and MODIS-derived subarctic Atlantic VGPM NPP estimates (Extended Data Figure 1), we spatially-composited (via averaging) monthly SeaWiFS and MODIS-Aqua sensor data over their years of common overlap (A.D. 2003-2007) to produce a single satellite VGPM-NPP product. Using this composited product we find generally positive trends in subarctic Atlantic NPP over the first decade of satellite monitoring, and generally negative trends over the subsequent (most-recent) decade (Extended Data Figure 2). However, it is noted that substantial spatial heterogeneity does occur. Note that sensitivity analyses pertaining to our decision to composite SeaWiFS and MODIS-Aqua derived VGPM estimates can be found in Extended Data Figure 2.

## S4. Processing of Continuous Plankton Recorder (CPR) survey data

A brief overview of the CPR survey, including the survey's data-collection methodology, potential biases (and associated adjustments), as well as limitations, can be found in the Methods portion of the main text (for an extensive review see ref. 53). Here, we provide specific detail on i) the availability of CPR survey data within the subarctic Atlantic (A.D. 1958-2016) and an intercomparison of summertime vs. annual averaging of CPR data, as well as ii) a comparison of data reduction techniques for estimating (spatially-integrated) subarctic Atlantic planktonic abundance trends from CPR data.

*Record availability by standard CPR region: summer vs. annual data*

We only considered data from CPR standard regions situated over the subarctic Atlantic (50-65˚N, 60-10˚W), limiting our analysis to the following 14 (out of the 41) standard CPR regions: A6, A8, B5-8, C5-8, D5-8 (ref. 53).

Because few years have data for all months out of the year in most subarctic Atlantic-situated standard regions, the convention for estimating interannual variability using CPR data is to average across years with ≥8 months of data (i.e., ≥2/3 of a year), though annual estimates can typically be improved when the remaining ≤4 months (i.e., ≤1/3 of a year) are first estimated prior to annual averaging[53]. Ref. 53 suggests the following method for estimating a missing month ($M$) of data:

$$M = \bar{M} * \frac{Y}{\bar{Y}} \qquad \text{eq. S1}$$

where $\bar{M}$ is the climatological monthly mean, and $Y$ is the annual mean which is normalized by $\bar{Y}$, the climatological annual mean. In Extended Data Fig. 3, we show the data availability in the subarctic Atlantic for the annual averaged CPR data (i.e., the percentage of years per standard region where number of months were ≥8) over the period A.D. 1958-2016.

We compare CPR data representing (conventionally) annually-averaged data to summertime-averaged data, that is, the mean for CPR data collected in the months (Apr-May-June-July-August-Sept). Following ref. 53, in this case we required ≥4 months (i.e., ≥2/3 of a summer) to compute the summertime average, whereby the missing summer months are estimated via eq. S1 (such that $Y$ now represents the annual summer mean and $\bar{Y}$ the climatological summer mean) prior to averaging. As can be seen in Extended Data Fig. 3a-b, withholding wintertime data facilitates marginal increases in the number of available years of data for most standard regions, particularly across the relatively poorly-sampled (albeit less-productive; Fig. 3a) central to western subarctic Atlantic.

We explore correlations of summertime- and annually-averaged CPR data to alternate subarctic Atlantic bioproductivity and climatological indices, reminiscent to the procedure highlighted in Extended Data Figure 8a. The results of the correlation analysis (not shown) reveal that the CPR correlations using annual-averages are highly similar to summertime averages, suggesting both approaches are reasonable for estimating long-term variability in CPR abundance data. In the following section, we extend our comparison of annually- vs. summertime averaged CPR time series to also compare methods of compositing time series between CPR standard regions.

## *Comparison of methods for determining CPR temporal variation*

The availability of 14 CPR standard regions for the subarctic Atlantic necessitated a spatially- and statistically- representative dimensional reduction technique to compute annual indices of PCI, diatom, dinoflagellate, and coccolithophore abundance, respectively. We considered three approaches: area-weighted averaging (AWA), inverse squared distance-infilling (ISD) averaging[17], and an empirical orthogonal function (EOF)-based statistical infilling method[42].

The AWA method provides the simplest approach, incorporating an area-weighted mean planktonic abundance estimate of all available CPR regions in a given year. Given that each CPR region entails varying amounts of missing data (e.g., due to year-to-year differences in shipping route coverage; see above section or ref. 53), some underlying (non-stationary) spatial bias may be introduced when estimating annual planktonic abundance with the AWA method. To help alleviate these potential biases, we explored ISD-infilling to first estimate missing years of data (e.g., ref. 17) prior to area-weighting averaging of the CPR regions' abundances. This approach relies on the physical rationale that planktonic abundances in CPR regions of closer proximity are, ostensibly, more closely-related than CPR regions farther afield. This deterministic data-infilling approach (i.e., ISD averaging) is contrasted by a probabilistic EOF-based infilling methodology[42]. Following ref. 17, for this procedure we first omitted CPR standard regions missing ≥20 years of data from the analysis, in order to remove spurious signals and thus improve convergence of the EOF-algorithm[42]. For the remaining CPR regions, EOF-based imputation of missing years of data[42] was conducted in a manner identical to that described in Methods (i.e., for the infilling of missing [MSA] data), prior to area weighted averaging.

A qualitative comparison of the annual time series resulting from all three compositing procedures can be found in Extended Data Fig. 3. Correlation analysis of both the summertime vs. annually time series for all three compositing techniques suggests that, in general, the EOF-data infilling technique provides the most internally consistent results (Table S2). Nonetheless, all three techniques show a notable degree of similarity across multidecadal timescales, suggesting that decadal-scale and longer productivity trends measured by the CPR survey are robust independent of which compositing approach is used. Due to its simplistic and intuitive approach, we highlight in Figure 4a and Extended Data Figure 8a of the main text CPR abundance time series using AWA.

## S5. Comparison of [MSA]-PC1 to regional sea ice behavior

Due to the reigning presupposition behind the use of ice core [MSA] as a proxy for sea ice extent[34,65], we compared trends in near-Greenlandic sea ice extent (SIE; defined as the summed area of sea-ice concentration grids that are ≥15%) against the [MSA]-PC1 time series. We used the updated National Snow and Ice Data Center (NSIDC) Historical Arctic Sea Ice gridded sea ice concentration product[77], which extends from A.D. 1850 to present and is available at monthly temporal resolution. We explored trends in SIE for both March and September, typically the months of maximum and minimum SIE in the Arctic, and across four distinct regions listed subsequently in order of decreasing presumed association to Greenland MSA deposition (see Section S1, above). Region 1 (60-70˚N, 315-340˚E) encompasses the vicinity of the Denmark Strait/Icelandic Basin, and is situated over the southeast margin of Greenland. This is the region over which the highest [MSA]-PC1 airmass transport probability density is situated (Fig. 1). Region 2 (60-85˚N, 315-15˚E) defines the entire eastern margin of Greenland extending west to Svalbard. Region 3 (45-85˚N, 280-315˚E) covers Baffin Bay, extending southward into the Labrador Sea beyond the southernmost mean SIE maximum reached over the period A.D. 1900-2013. Region 4 comprises the sum of Region 2 and 3, capturing both eastern and western Greenlandic sea-ice behavior.

As a rough test of causality (i.e., assuming *a priori* that annual production and the ease of subsequent airborne deposition of MSA onto the Greenland Ice Sheet is in some way modulated by the extent of sea ice proximal to Greenland[65]), we computed the lag-0 ordinary least squares linear regression of annual [MSA]-PC1 to regional SIE in both September and March over four different tests: Test 1) the satellite-era correlations (1979-2009; $n$ = 31 years; representing the best-resolved period of satellite sea-ice observation); Test 2) the long term *non-detrended* annual-scale correlations (1850-2009; $n$ = 160); Test 3) the long term *linearly-detrended* annual-scale correlations (1850-2009; $n$ = 160; note that both [MSA]-PC1 and regional SIE were linearly-detrended prior to regression analysis); Test 4) correlations following 10-yr lowpass (Butterworth) filtering of the time series (1850-2009; $n$ = 160 decades). Results of the four tests, provided in full in Table S1, show that only 2/32 tests, both for Region 3 in September, resulted in a significant [MSA]-PC1 – SIE correlation ($p \leq 0.05$) when adjusting for serial correlation amongst the paired time series[54].

In addition to the correlation analyses, we point out two additional qualitative discrepancies between [MSA]-PC1 and SIE (not shown). First, multidecadal-scale phasing between [MSA]-PC1 and SIE does not appear stationary for any of the four SIE regions. For example, the generally positive association between SIE and [MSA]-PC1 trends during the earlier half of the sea-ice satellite-observational period (1979 to mid-1990's) is not preserved into present. For example, since A.D. 1979, summertime and wintertime SIE has declined at an accelerating rate[73], while the [MSA]-PC1 series shows a generally-increasing trend. Second, a conspicuous increase in SIE found across all four regions during the late 1960's to early 1970's (reflecting the Great Salinity Anomaly, a period of elevated Arctic-basin sea ice export into the Barents/Greenlandic Seas[78]), does not have an appropriate analogue in the [MSA]-PC1 series.

Overall, our analysis suggests SIE variability is not the primary signal captured by [MSA] variations across interior Greenland, in contrast to prior suggestions[34]. While the use of [MSA] as a sea ice proxy appears to be robust for records from low-lying ice caps in Svalbard[79], situated at the summertime ice marginal extent of the Barents Sea, we argue that the leading mode of interior-Greenlandic [MSA] variability more likely reflects spatially-integrated variations in DMS production centered around the central to northeastern subarctic Atlantic, a region largely removed from the summertime sea ice margin.

## S6. NOAA Global Surface Seawater DMS Database measurements

In Extended Data Figure S7b, we plot a global comparison of [$DMS_{SW}$] to VGPM-derived NPP. The binning and regression methodology is described in the Methods of the main text. The results clearly show that [$DMS_{SW}$] variations within the subarctic Atlantic (Extended Data Figure 7a reproduced from Fig. 3b in the main text) capture a much larger fraction of NPP variability (51% using an OLS regression with $n$ = 224 points; 57% using a WLS regression with $n$ = 186 points (where the difference in $n$ between the two regressions discriminates between 1˚x1˚x1-month binned [$DMS_{SW}$] values representing >1 measurement) than exhibited on a global scale ($r$ = 0.32 and $r$ = 0.33 for the WLS and OLS, respectively).

Due to the large number data used in the global regression ($n$ = 3045 observations for OLS, $n$ = 2221 observations for WLS), both the WLS and OLS regressions for global [$DMS_{SW}$]-NPP data are highly significant ($p \ll 0.0001$) despite the substantially smaller Pearson $r$ values. To adjust for this potential statistical artifact, a large number (10,000) of global [$DMS_{SW}$]-NPP WLS regressions were conducted, whereby for each iteration $n$ = 186 samples were randomly drawn (with replacement) from the $n$ = 2221 global WLS [$DMS_{SW}$]-NPP pairings in order to approximate the degrees of freedom used in the subarctic Atlantic WLS regression. The results of this procedure indicated a median $r$ = 0.33, and 99$^{th}$ percentile of $r$ = 0.56, indicating the globally-integrated [$DMS_{SW}$]-NPP relationship to be less robust than in the subarctic (Extended Data Figure 7c).

## S7. Defining the subarctic Atlantic "warming hole" (SST) and calculation of the observational AMOC index

Broad portions of the observational subarctic Atlantic SST (or, near-equivalently, SAT) record are characterized by long term cooling in spite of global long-term warming[6-7,80-81]. Due to the conspicuous nature of this so-called Atlantic "warming hole" and its relationship to both atmospheric forcing[81], lateral convection[22], and, across longer timescales, thermohaline overturning strength[6], we target this region as an indicator for linking primary productivity trends within the context of subarctic Atlantic climate dynamics. Trends in sea-surface temperatures (SST) were first analyzed using the ERSST (v5; ref. 30) and HadISST- (v1.1; ref. 58) mean-monthly reanalysis datasets over the period A.D. 1870-2016. We defined grid cells showing a linear decrease over this 146-yr period within the subarctic Atlantic (50-65˚N, 60-10˚W) as the Atlantic warming hole region. Despite some discrepancies between the spatial extent of the warming hole between the two datasets – likely a result of differences in grid-cell resolution (2˚x2˚ for ERSSTv5; 1˚x1˚ for HadISSTv1.1) – both area-weighted mean annual SST anomalies reveal a high degree of consistency (bottom panel; $r$ = 0.91; Extended Data Figure 9), indicating temperature covariability across the subarctic Atlantic is generally coherent between datasets.

Following ref.'s 6-7, we used the Atlantic warming hole region to calculate the observational AMOC index from A.D. 1870-2016 as the difference between the mean annual warming hole SST anomaly and the mean annual northern hemisphere SST anomaly. As shown in the supplementary information of ref. 6, primary features of the AMOC index are preserved when using either SST (i.e., only oceanic grid points) or SAT (i.e., both oceanic and continental grid points) as the base unit. Similar to the warming hole comparison above, we find minor differences between the ERSSTv5 derived- and the HadISSTv1.1 derived AMOC indices. Further, both indices – following 10-yr lowpass filtering – show strong (Pearson product-moment) correlation with the (10-yr lowpass filtered) reconstructed AMOC index of ref. 6 (A.D. 1870-1995.; $r_{ERSST}$ = 0.81; $r_{HadISST}$ = 0.82; Extended Data Fig. 9). Due to differences in base unit (SAT vs. SST) and reference datasets used, small magnitudinal offsets between the observed SST AMOC index and the reconstructed SAT AMOC index of ref. 6 do exist. As such, the observational ERSSTv5 AMOC index provided in Figure 3a of the main text was bias corrected and adjusted to the reconstructed SAT AMOC index of ref. 6 to improve visual clarity. The adjustment does not affect the correlation analyses listed above or within the main text.

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# III. Supplementary Tables

Table S1. Explorative correlation analysis of [MSA]-PC1 to SIE in regions proximal to Greenland. Values are the correlation values (Pearson *r*) of [MSA]-PC1 with March SIE (~maximum extent), while values in parentheses represent September (~minimum extent) SIE correlations. Bolded values represent significance at the $p < 0.05$ confidence level following a Monte Carlo Fourier-phase randomization procedure (10,000 tests per pairing), used to empirically account for serial correlation amongst paired time series[54].

| | **Region 1** | **Region 2** | **Region 3** | **Region 4** |
|---|---|---|---|---|
| **Test 1: Annual satellite period (A.D. 1979-2009; *n* = 31)** | 0.03 (-0.23) | -0.05 (-0.08) | -0.08 (-0.02) | -0.09 (-0.06) |
| **Test 2: Annual reanalysis period (A.D. 1850-2009; *n* = 160)** | 0.06 (0.04) | 0.25 (0.17) | 0.03 (**0.37**) | 0.21 (0.31) |
| **Test 3: Annual, linearly detrended (A.D. 1850-2009; *n* = 160)** | -0.11 (-0.07) | -0.11 (-0.06) | 0.05 (-0.00) | 0.06 (-0.04) |
| **Test 4: 10-yr lowpass filtered (A.D. 1850 – 2009; *n* = 16)** | 0.08 (0.11) | 0.31 (0.29) | -0.04 (**0.58**) | 0.28 (0.46) |

Table S2. Linear correlation analysis of three methods of CPR-based compositing for i) diatom, ii) dinoflagellate, and iii) coccolithophore abundances, comparing both summer vs. annual averaging. Numbers represent Pearson product-moment correlations ($r$; A.D. 1958-2016), with Monte Carlo-based[54] significance level in parentheses. A significance level of "$<10^{-4}$" represents an observed $r$-value whose magnitude was greater than (10,000) correlations created using pseudo-random surrogate data (see Methods).

| **Functional group:** | **i) Diatom** | | | **ii) Dinoflagellate** | | | **iii) Coccolithophore** | | |
|---|---|---|---|---|---|---|---|---|---|
| **Summer AWA** | 0.91 $(2.1\times10^{-4})$ | 0.30 (0.19) | 0.68 $(1.5\times10^{-2})$ | 0.83 $(4.7\times10^{-4})$ | 0.06 (0.40) | 0.66 $(6.0\times10^{-4})$ | 0.63 (0.10) | 0.77 $(5.4\times10^{-2})$ | 0.74 $(7.5\times10^{-2})$ |
| **Summer ISD** | 0.50 $(2.4\times10^{-2})$ | 0.96 $(<10^{-4})$ | 0.84 $(<10^{-4})$ | 0.17 (0.28) | 0.94 $(<10^{-4})$ | 0.38 $(6.6\times10^{-2})$ | 0.69 $(7.2\times10^{-2})$ | 0.82 $(2.8\times10^{-2})$ | 0.80 $(4.6\times10^{-2})$ |
| **Summer EOF** | 0.77 $(4.6\times10^{-3})$ | 0.78 $(<10^{-4})$ | 0.98 $(<10^{-4})$ | 0.69 $(2.9\times10^{-4})$ | 0.42 $(4.0\times10^{-2})$ | 0.96 $(<10^{-4})$ | 0.81 $(2.9\times10^{-2})$ | 0.89 $(1.0\times10^{-2})$ | 0.99 $(<10^{-4})$ |
| | **Annual AWA** | **Annual ISD** | **Annual EOF** | **Annual AWA** | **Annual ISD** | **Annual EOF** | **Annual AWA** | **Annual ISD** | **Annual EOF** |

Table S3. Overview of climatic indices, reanalysis, and biological data used.

| Variable | Dataset name | URL (as of Feb. 2018) | Time period covered | Resolution | Regridding performed? | Citation |
|---|---|---|---|---|---|---|
| Sea surface temperature (SST) | NOAA ERSST (v5) | https://www.esrl.noaa.gov/psd/data/gridded/data.noaa.ersst.html | Jan 1854 – (monthly) | 2˚ | N | 30 |
| | HadISST (v1.1) | https://www.metoffice.gov.uk/hadobs/hadisst/ | Jan 1870 – (monthly) | 1˚ | N | 58 |
| Sea-ice extent (SIE) | NSIDC Gridded Historical Sea Ice (v1) | https://nsidc.org/data/g10010 | Jan 1850 – Dec. 2013 (monthly) | 0.25˚ | N | 73 |
| Dimethylsulfide sea-surface concentration | NOAA Global Surface Seawater DMS Database | https://saga.pmel.noaa.gov/dms/ | 1972 – (Inconsistent sampling) | n/a | Y (1˚ CPR standard region-wise binning) | 23,52 |
| Phytoplankton Color Index (PCI), Diatom, Dinoflagellate, & Coccolithophore abundance | SAHFOS Continuous Plankton Recorder (CPR) survey | https://www.sahfos.ac.uk/ | 1958 – (monthly) | n/a | n/a | 53 |
| North Atlantic Oscillation (NAO) index | CRU Station-based | https://crudata.uea.ac.uk/cru/data/nao/ | n/a | n/a | n/a | 24 |
| | NAO – proxy based reconstruction | https://www1.ncdc.noaa.gov/pub/data/paleo/contributions_by_author/ortega2015/ortega2015nao.txt | 1767 – 1969 | n/a | n/a | 25 |
| Subpolar Gyre (SPG) Index | Merged MICAM (model) and AVISO SSH (observed) | Observed: https://data.marine.gov.scot/dataset/sub-polar-gyre-index; model: received upon request from H. Hatun, Feb. 2018 | 1960-2003 (modeled; ref. 22); 1993-2012 (observed; ref. 82); $r$ = 0.96 for 11 years of overlap; A.D. 1993-2003). Note that the observed series was truncated due to recent, ongoing discussion on defining the SPG index across recent years[83]. | n/a | n/a | 22; 82 |
| AMOC Index | AMOC – proxy based reconstruction | http://www.pik-potsdam.de/%7Estefan/amoc_index_data.html | 1767 – 1995 | n/a | n/a | 7 |
| Dimethylsulfide monthly climatology | SOLAS Project | https://www.bodc.ac.uk/solas_integration/implementation_products/group1/dms/ | n/a (monthly climatology) | 1˚ | N | 23 |
| Ocean Net Primary Productivity | Vertically Generalized Production Model (VGPM) using SeaWiFS-r2018 reprocessing | http://orca.science.oregonstate.edu/1080.by.2160.monthly.hdf.vgpm.s.chl.a.sst.php | Oct. 1997 – Dec. 2007 (monthly) | 1/6˚ | Y (1˚) | 12 |
| | Vertically Generalized Production Model (VGPM) using MODIS-r2018.0 (GSM) reprocessing | http://orca.science.oregonstate.edu/1080.by.2160.monthly.hdf.vgpm.m.chl.m.sst.php | Jul. 2002 – Dec. 2017 (monthly) | 1/6˚ | Y (1˚) | 12 |
| | Carbon-based Productivity Model using GSM-v8; SeaWiFS-r2018 | http://orca.science.oregonstate.edu/1080.by.2160.monthly.hdf.cbpm2.s.php | Oct. 1997 – Dec. 2007 (monthly) | 1/6˚ | Y (1˚) | 57 |

| | | | | | | |
|---|---|---|---|---|---|---|
| | reprocessing | | | | | |
| | Carbon-based Productivity Model using MODIS-r2018.0 reprocessing (GSM) | http://orca.science.oregonstate.edu/1080.by.2160.monthly.hdf.cbpm2.m.php | Jul. 2002 – Dec. 2017 (monthly) | 1/6˚ | Y (1˚) | 57 |