Industrial-era decline of subarctic Atlantic productivity

Matthew B. Osman[1*], Sarah B. Das[2], Luke D. Trusel[3], Matthew J. Evans[4], Hubertus Fischer[5], Mackenzie M. Grieman[6], Sepp Kipfstuhl[7], Joseph R. McConnell[8], Eric S. Saltzman[6]

1. Massachusetts Institute of Technology/Woods Hole Oceanographic Institution Joint Program in Oceanography/Applied Ocean Sciences and Engineering, Woods Hole, MA, USA
2. Department of Geology and Geophysics, Woods Hole Oceanographic Institution, Woods Hole, MA, USA
3. Department of Geology, Rowan University, Glassboro, NJ, USA
4. Department of Chemistry, Wheaton College, MA, USA
5. Climate and Environmental Physics, Physics Institute and Oeschger Centre for Climate Change Research, University of Bern, Bern, Switzerland
6. Department of Earth System Science, University of California, Irvine, Irvine, California, USA
7. Alfred-Wegener-Institute, Bremerhaven, Germany
8. Division of Hydrologic Sciences, Desert Research Institute, Reno, USA

* Email: osmanm@mit.edu

Marine phytoplankton play a critical role in modulating marine-based food webs[1], fishery yields[2], and the global drawdown of atmospheric CO_2 (ref. 3). Due to sparse measurements prior to 21st century satellite monitoring, however, little is known of the long-term response of planktonic stocks to climate forcing. Here we produce the first continuous, multi-century record of subarctic Atlantic marine productivity, showing a marked 10 ± 7% decline has occurred across this highly-productive ocean basin over the last two centuries. We support this conclusion through the application of a novel marine-productivity proxy, established using a unique signal of planktonic-derived aerosol commonly identified across an array of Greenlandic ice cores. Utilizing contemporaneous satellite-era observations, we demonstrate this signal's use as a robust and high-resolution proxy for spatially-integrated marine productivity variations. We show that the initiation of declining subarctic Atlantic productivity broadly coincides with the onset of Arctic surface warming[4], and that productivity strongly covaries with regional sea-surface temperatures and basin-wide gyre circulation strength over recent decades. Taken together, our results suggest the industrial-era productivity decline may be evidence of the predicted[5] collapse of northern Atlantic planktonic stocks in response to a weakened Atlantic Meridional Overturning Circulation

(AMOC)[6-8]. Continued AMOC weakening, as projected for the 21st century[9,10], may therefore result in further productivity declines across this globally-relevant region.

The subarctic Atlantic (50-65˚N, 60-10˚W) comprises one of the world's most biologically productive seasonal phytoplankton blooms[11-14]. Bloom magnitude varies annually, in response to controls such as the timing and abundance of light and nutrients in the upper ocean[12-13], and predator-prey coupling dynamics[14]. These biophysical controls, in turn, vary in response to underlying physical drivers that are sensitive to long-term changes in upper-ocean climatic forcing, such as the mixed layer depth (MLD), sea-surface temperature (SST), baroclinicity, and wind.

Over the preceding ~200 years (i.e. the industrial-era) the northern Atlantic has undergone numerous climatic perturbations outside the range of naturally forced variability, resulting in widespread surface warming[4], AMOC slowdown[6-8], sea ice decline[15], and accelerating Greenland Ice Sheet (GrIS) runoff[16]. Contemporaneous estimates of primary productivity are less well resolved. Satellite-derived planktonic biomass concentrations, extending back only to late-1997 (and intermittently to 1979; ref. 11-12), do not reveal significant decadal-scale trends, but rather, modest productivity variability over the first two decades of 21st century monitoring (Extended Data Fig.'s 1 and 2; Supplementary). Earlier spatiotemporally sparse sources, including ship-based ocean color[11] and planktonic abundance observations[17] (Extended Data Fig. 3; Supplementary), however, hint at a longer-term, 20th century decline. To date, no spatially-reconciled, temporally-resolved reconstruction of basin-scale primary productivity exists over the pre- to post-industrial transition. This limits our ability to quantify climatic impacts on subarctic Atlantic ecosystems, and contextualize model-based predictions of future ecologic responses to anthropogenic forcing[9].

Here we use records of a marine-derived biogenic aerosol (methanesulfonic acid concentration, [MSA]) from GrIS ice cores to reconstruct annual subarctic Atlantic productivity variability over the preceding ~two and a half centuries. At high latitudes, MSA is solely produced as an oxidative byproduct from oceanic dimethylsulfide (DMS) emissions[18]. DMS, in turn, is linked to several planktonic life-cycle processes involving dinoflagellate, haptophyte (including coccolithophores) and, to a lesser extent, diatom, chrysophyte, and prasinophyte assemblages[19]. Notwithstanding uncertainties in the long-term feedbacks between DMS emissions and climate[20], we demonstrate that GrIS-[MSA] records provide a first order proxy for regional ocean-atmosphere fluxes of DMS[18] (Methods), which are tightly coupled to changes in nearby marine productivity[21].

We combine 12 high-resolution [MSA] records (Methods) to examine the covariation of [MSA] across the GrIS (Extended Data Table 1). To physically constrain potential DMS emission source regions across the 12 ice core sites, we conduct daily atmospheric back-trajectory analyses during the time of MSA summertime maximum, June-July-August (JJA; Extended Data Table 1), over a multi-decadal time-frame (AD 1948-2013; Methods). A conspicuous multi-century decline in [MSA] is evident in nearly all records (Fig. 1a), irrespective of their GrIS locations and primary summertime airmass-trajectory pathways – trending from predominantly southeasterly-dominated (Irminger Sea origin) atmospheric influence at southern-situated GrIS sites to southwesterly-dominated (Labrador Sea origin) influence at northern-situated sites (Fig. 1b; Extended Data Figures 4-5). Moreover, when averaging the 12 records into either Irminger Sea- ($n = 7$ sites; Fig. 1b) or Labrador Sea-dominated constituents ($n = 5$ sites), we find significant GrIS-[MSA] covariation down to annual-timescales ($p < 0.0001$; Extended Data Figure 6; Supplementary).

This strong coherence suggests that time-variability of MSA deposition across the GrIS is dominated by a common, large-scale mode of North Atlantic DMS production and emissions. Using a probabilistic principal component analysis (PCA) methodology, we extract this common signal of GrIS-[MSA] variability over the period A.D. 1767-2013 (Methods). The leading principal component, [MSA]-PC1, robustly captures the multi-century decline in industrial-era [MSA] observed across the individual records, while significantly ($p = 0.002$) explaining nearly half of the GrIS-[MSA] variability (median/mode at ~44%; Fig. 2a). Correlation of the [MSA] records to the resultant [MSA]-PC1 signal furthermore reveals each record to be significantly and positively related ($p < 0.1$; Methods; Extended Data Table 1), with centrally-situated GrIS sites exhibiting the strongest covariation with [MSA]-PC1 (Fig. 2a). Exploiting this spatial-loading pattern, we statistically-composite the 12 sites' back-trajectory results (Methods; Extended Data Figures 4-5) underscoring the west-central to northeastern (NE) subarctic Atlantic basin as the most probable source of GrIS-deposited MSA (Fig. 2b-c). Significantly, this MSA source region, centered over the highly-productive[1,12,17] and climatically-sensitive[1,6-8,22] Irminger and Labrador Seas proximal to the upwind Greenland coast, overlaps with the greatest Atlantic-sector surface JJA DMS seawater concentrations[23], [DMS_{SW}] (and, relatedly, DMS-emissions; Fig 3a).

Strong spatial coherence further exists between summertime [DMS_{SW}] and satellite-derived estimates of net primary productivity (NPP) from the chlorophyll-α (Chl-α) dependent Vertically Generalized Production Model[12] (VGPM; Supplementary) across the northern Atlantic sector, despite differences in the collection and spatial scalability of these two data sources (Methods; Fig. 3a). The observed similarity suggests that past variations in northern Atlantic DMS production, as inferred from [MSA]-PC1, also provide a signal of past productivity variations across this sector, given time-averaged scalability in the ocean-atmosphere emission rate of DMS to DMS_{SW} production[23]. Indeed, correlation of VGPM-derived NPP against [DMS_{SW}] measurements from the NOAA Global Surface Seawater DMS Database[23] (Methods) supports a strong spatiotemporal association ($r = 0.75$) between subarctic Atlantic productivity and the magnitude of DMS production (Fig. 3b), a relationship well-above globally-integrated values (Extended Data Fig. 7c; Supplementary). A similar regression analysis against independently-derived counts of DMS-producing diatom, dinoflagellate, and coccolithophore relative abundance from the Continuous Plankton Recorder (CPR) ship-survey (Methods) also yields significant relationships with [$\mathrm{DMS_{SW}}$] in all three functional groups ($p < 0.005$; Extended Data Fig. 7d-f), implying that variations in DMS production and emissions across the subarctic Atlantic are well representative of contemporaneous broad-scale changes in planktonic biomass and productivity.

The above back-trajectory and correlation analyses are connected through an empirical orthogonal function (EOF) analysis of satellite NPP, which reveals the leading mode of summertime and annual NPP-variability (NPP-PC1; ~20% and 24% explained-variance, respectively) to be closely-aligned with [MSA]-PC1 over multiannual timescales (Fig. 3d; Methods), while also remaining notably consistent with spatially-integrated summertime and annual NPP yields ($r = 0.75$ and 0.91, respectively; Extended Data Fig. 2). This latter similarity is underscored in Figure 3c by the broadly-coherent loading pattern of NPP-PC1 (EOF1), spatially-linking marine productivity across a broad portion of the Irminger, Labrador, as well as western-Icelandic Seas (Fig. 3c). Importantly, the analysis also reveals the extrema of NPP-EOF1 to directly overlie the [MSA]-PC1 airmass density maxima and altitude minima of Figures 2b and 2c, respectively. Provided moderate stability in the spatial character of productivity variability during the past (and the underlying phytoplankton assemblages comprising it), this independently confirms that DMS-emissions from this region, once converted to MSA in the atmosphere and deposited atop the GrIS, are ideally suited for reconstructing broad-scale subarctic Atlantic productivity variations.

Results from our combined back-trajectory (Fig. 2b-c), correlation (Fig. 3b; Extended Data Fig. 7), and EOF (Fig. 3c) analyses support our use of the [MSA]-PC1 signal (Fig. 2a) as an index for past marine productivity variations across the subarctic Atlantic basin. Our ice core-based productivity index is remarkably consistent with the 20th century decline in (basin-scale) North Atlantic planktonic stocks previously reported by ref. 11 ($p < 0.0001$; Extended Data Figure 8a), as well as broadly congruent with several CPR-based indices of subarctic Atlantic planktonic abundance (Fig. 4a; Methods; Extended Data Fig. 3). Notably, all records show a pronounced decline over the second half of the 20th century, followed by recent intermittent (likely natural-decadal) productivity variability that has so-far characterized the contemporary satellite-era (Fig. 4a; Extended Data Fig. 2). Moreover, our new multi-century productivity record significantly extends prior spatiotemporally-limited ship-based observations beyond the mid-20th century[11], suggesting the 20th century decline is part of a much longer-term trend.

The additional temporal context of our productivity index allows us to investigate subarctic Atlantic productivity responses to changes in atmospheric and oceanic forcing over recent decades, here characterized by indices of the North Atlantic Oscillation[24-25] (NAO) and Subpolar Gyre (SPG) circulation strength[22]. Using correlation analysis (Extended Data Figure 8a; Methods), we find the NAO is only weakly related to our reconstructed bioproductivity variations while, in contrast, SPG strength indicates significant negative influence over decadal timescales as previously indicated from sparse ship-based color data[17]. Modeling studies suggest that during weakened[26-27] and (or) contracted[22] SPG states, wintertime MLD's deepen across the central-NE subarctic Atlantic and shoal across the Labrador Seas[27]. Thus, in addition to its first-order inverse effect on NE Atlantic SST variability[22] (Fig. 4a), this could explain how a weak SPG, by enhancing wintertime deep-water nutrient replenishment to the euphotic zone[1,5,9,13], or by delaying the seasonal-onset of predatorial grazing cycles[14] across the ecologically-productive central-NE Atlantic[17] (Fig. 3a), could lead to increases in NPP as observed by our results.

Differential change-point analysis[4,16] of our [MSA]-PC1 record suggests declining subarctic Atlantic productivity began in A.D. 1816 ± 11 years (Fig. 4a; Methods), broadly consistent with the onset of regional surface temperature warming[4]. Applying a calibration derived from the relationship between [MSA]-PC1 and the leading mode of 21st century satellite NPP (Fig. 3d; Extended Data Fig. 2), we calculate an estimated ~10 ± 7% decline ($\pm 2\sigma$; Fig. 4b) in contemporary subarctic Atlantic NPP yields since the industrial-era onset. Despite the uncertainties of this estimate, arising from the short time-span of satellite NPP estimation (Fig. 4a) and limited GrIS-[MSA] data-availability during this period (Fig. 3d; Methods), the onset of declining subarctic Atlantic productivity appears temporally-consistent with the (~15%) decline in industrial-era Atlantic thermohaline overturning strength (i.e., AMOC) recently inferred from Labrador and Irminger basin marine sediments[8]. We similarly observe strong multidecadal- to centennial-scale correspondence ($p < 0.0001$) between our productivity index and a separate, high-resolution terrestrial proxy-based reconstruction[6] of AMOC predicated upon NE Atlantic upper ocean heating anomalies (Fig. 4a; Extended Data Fig. 9; Supplementary). These results, suggesting a positive relationship between productivity, subarctic Atlantic SSTs and large-scale thermohaline variability across decadal-scale and longer timescales, contrast model-based contentions of a positive (i.e., reinforcing) influence of SPG-circulation strength on both subarctic Atlantic overturning[26] and, by extension, productivity.

The strong observed coherence between productivity and AMOC strength, moreover, supports a previous model-based hypothesis[5] that a sustained, industrial-era slowdown of AMOC[6-8] would lead to dramatically reduced planktonic yields across the northern Atlantic. In particular, both [MSA]-PC1 and AMOC exhibit corresponding multicentury-scale[6,8] lows during the 1980's to 1990's. This time period coincides with a massive accumulation of freshwater (~15,000 km^3 from 1965-1990) into the subarctic Atlantic basin following the Great Salinity Anomaly of the late-1960's (ref. 28; Fig. 4a). According to the relationships illuminated by our results, the decrease in upper-ocean densities associated with this event, hypothesized to have weakened deep-water formation across the Labrador and Irminger Seas[6,28], may also have led to a diminishing of northern Atlantic planktonic stocks, presumably either through long-term shoaling of wintertime MLDs and the gradual diminishing of euphotic nutrient concentrations[1,5,9], or through first-order thermal influences[1,12]. Further, the onset of industrial-era Arctic sea ice decline and elevated GrIS runoff, commencing several decades after our productivity decline and accelerating into present[15,16], suggests that a long-term freshening of NE subarctic Atlantic surface waters – similarly implicated in driving the industrial-era AMOC decline[1,6-8] – may have contributed in sustaining the industrial-era productivity decline over the late 19th and 20th centuries. Clearly, more work is needed to understand these complex relationships.

Our ice core based index of subarctic Atlantic bioproductivity highlights the sensitivity of marine-based autotrophic-ecosystems to industrial-era forcing and provides context for projected future ecologic changes[6]. Although previous ship-based[11,17] and satellite-derived reconstructions suggested an early 21st century reversal in the 20th century subarctic Atlantic productivity decline (Fig. 4a), results from our [MSA]-PC1 proxy show the decline on which this intermittent 21st century increase is superimposed is much longer than previously observed[11], and may still be ongoing. Monitoring of the AMOC at 26.5˚N since 2004 has shown a decade-long decline in meridional heat transport, decreasing as much as 10 times faster than model-predicted slowdowns[29]. Given the multiyear time-lag required of Atlantic-wide mixing[13,22], as well as the ongoing, nonlinear rise in Greenland runoff[16] believed to contribute to subarctic Atlantic freshening and AMOC slowdown over multidecadal to centennial timescales[6,8,10], we speculate declining subarctic Atlantic productivity will characterize the coming decades with important implications on future atmospheric carbon drawdown[3] and northern Atlantic fisheries[2].

References

1. Barton, A. D., Irwin, A. J., Finkel, Z. V & Stock, C. A. Anthropogenic climate change drives shift and shuffle in North Atlantic phytoplankton communities. *Proc. Natl. Acad. Sci.* **113,** 2964–2969 (2016).
2. Stock, C. A. *et al.* Reconciling fisheries catch and ocean productivity. *Proc. Natl. Acad. Sci.* **114,** E1441–E1449 (2017).
3. Takahashi, T. *et al.* Climatological mean and decadal change in surface ocean pCO2, and net sea-air CO_2 flux over the global oceans. *Deep. Res. Part II Top. Stud. Oceanogr.* **56,** 554–577 (2009).
4. Abram, N. J. *et al.* Early onset of industrial-era warming across the oceans and continents. *Nature* **536,** 411–418 (2016).
5. Schmittner, A. Decline of the marine ecosystem caused by a reduction in the Atlantic overturning circulation. *Nature* **434,** 628–633 (2005).
6. Rahmstorf, S. *et al.* Exceptional twentieth-century slowdown in Atlantic Ocean overturning circulation. *Nat. Clim. Chang.* **5,** 475–480 (2015).
7. Caesar, L., Rahmstorf, S., Robinson, A., Feulner, G. & Saba, V. Observed fingerprint of a weakening Atlantic Ocean overturning circulation. *Nature* **556,** 191–196 (2018).
8. Thornalley, D. J. R. *et al.* Anomalously weak Labrador Sea convection and Atlantic overturning during the past 150 years. *Nature* **556,** 227–230 (2018).
9. Moore, J. K. *et al.* Sustained climate warming drives declining marine biological productivity. *Science.* **359,** 1139–1143 (2018).
10. Bakker, P. *et al*. Fate of the Atlantic Meridional Overturning Circulation: strong decline under continued warming and Greenland melting. *Geophys. Res. Lett*. **43**, 12252–12260 (2016).
11. Boyce, D. G., Lewis, M. R. & Worm, B. Global phytoplankton decline over the past century. *Nature* **466,** 591–596 (2010).
12. Behrenfeld, M. J. & Falkowski, P. G. Photosynthetic rates derived from satellite-based chlorophyll concentration. *Limnol. Oceanogr.* **42,** 1–20 (1997).
13. Sverdrup, H. U. On conditions for the vernal blooming of phytoplankton. *ICES J. Mar. Sci.* **18,** 287–295 (1953).
14. Behrenfeld, M. J. & Boss, E. S. Resurrecting the Ecological Underpinnings of Ocean Plankton Blooms. *Ann. Rev. Mar. Sci.* **6,** 167–194 (2014).
15. Kinnard, C. *et al.* Reconstructed changes in Arctic sea ice over the past 1,450 years. *Nature* **479,** 509–512 (2011).
16. Trusel, L. *et al.* Nonlinear rise in Greenland runoff in response to post-industrial Arctic warming, *Nature* **564**, 104-108 (2018).
17. Hátún, H. *et al.* Large bio-geographical shifts in the north-eastern Atlantic Ocean From the subpolar gyre, via plankton, to blue whiting and pilot whales. *Progr. in Oceanogr.* **80**, 149-162 (2009).
18. Legrand, M. *et al.* Sulfur-containing species (methanesulfonate and SO_4) over the last climatic cycle in the Greenland Ice Core Project (central Greenland) ice core. *J. Geophys. Res. Ocean.* **102,** 26663–26679 (1997).
19. Stefels, J., Steinke, M., Turner, S., Malin, G., & Belviso, S. Environmental constraints on the production and removal of the climatically active gas dimethylsulphide (DMS) and implications for ecosystem modelling, *Biogeochemistry* **83**, 245-275 (2007).
20. Charlson, R. J., Lovelock, J. E., Andreae, M. O. & Warren, S. G. Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate. *Nature* **326,** 655–661 (1987).
21. Polashenski, D. J., *et al*. Denali ice core methansulfonic acid records North Pacific marine primary production, *J. Geophys. Res. Atmos.*, **123**, 4642-4653 (2018).
22. Hátún, H., Sande, A. B., Drange, H., Hansen, B. & Valdimarsson, H. Influence of the Atlantic subpolar gyre on the thermohaline circulation. *Science.* **309,** 1841–1844 (2005).
23. Lana, A. *et al.* An updated climatology of surface dimethlysulfide concentrations and emission fluxes in the global ocean. *Global Biogeochem. Cycles* **25,** 1–17 (2011).

24. Jones, P. D., Jónsson, T., Wheeler, D., Extension to the North Atlantic Oscillation using early instrumental pressure observations from Gibraltar and South-West Iceland. *Int. J. Climatol.* **17**, 1433-1450 (1997).
25. Ortega, P. *et al*. A model-tested North Atlantic Oscillation reconstruction for the past millennium. *Nature* **523,** 71–74 (2015).
26. Sgubin, G., Swingedouw, D., Drijfhout, S., Mary, Y. & Bennabi, A. Abrupt cooling over the North Atlantic in modern climate models. *Nat. Comm*. **8**, 14375 (2017).
27. Born, A. *et al*. Is the Atlantic subpolar gyre bistable in comprehensive coupled climate models? *Clim. Dyn.* **40,** 2993-3007 (2013).
28. Curry, R. & Mauritzen, C. Dilution of the northern North Atlantic Ocean in recent decades. *Science* **308***, 1772-1774* (2005).
29. Srokosz, M. A. & Bryden, H. L. Observing the Atlantic Meridional Overturning Circulation yields a decade of inevitable surprises. *Science* **348,** (2015).
30. Huang, B. *et al.* Extended reconstructed Sea surface temperature, Version 5 (ERSSTv5): Upgrades, validations, and intercomparisons. *J. Clim.* **30,** 8179–8205 (2017).

Acknowledgements

We thank David Thornalley, Rubao Ji, and Øyvind Paasche for their thoughtful feedback on early versions of the study, as well as Hjálmar Hátún, Daniel G. Boyce, and David Johns for contributing oceanographic and planktonic data shown in the study. We thank Michel Legrand for sharing the GRIP93a ice core [MSA] data. Funding for retrieval and analysis of the GC ice core was provided by the US National Science Foundation (NSF) Arctic System Science Program (award OPP-1205196 to S.B.D. and award OPP-1205008 to M.J.E.). M.B.O. acknowledges support from the U.S. Department of Defense Office of Naval Research – National Defense Science and Engineering Graduate (NDSEG) fellowship, an Ocean Outlook Fellowship to the Bjerknes Centre for Climate Research (University of Bergen, Norway). M.B.O. and S.B.D. further acknowledge institutional support from the Woods Hole Oceanographic Institution (WHOI). H.F. gratefully acknowledges support of the tracer glaciological work along the North Greenland Traverse (NGT) 1993-95 by the German Research Foundation (DFG) and the long-term support of ice core science at the University of Bern by the Swiss National Science Foundation (SNF). Collection and analysis of the D4, Tunu, and Summit2010 cores were supported by NSF grants 0221515, 0909541, and 1204176 to JRM. We thank the students and staff of the DRI ice core group for assistance in the field and lab. E.S.S. acknowledges NASA grant NNX15AF31G. We thank the NSF Ice Core Facility (formerly NICL), Ashley York, Matt Bingham, Madeline Hatch, and Milton Academy students for GC ice core sampling, processing, and [MSA] analysis support.

Author contributions

M.B.O. conceived of and designed the study with input from S.B.D. S.B.D, M.B.O., and L.D.T collected the GC ice core. M.J.E. analyzed the GC ice core chemistry. H.F. and S.K. led the collection and chemical analyses of all five NGT records. M.G., J.R.M., and E.S.S. jointly conducted the D4 and TUNU chemical analyses. J.R.M. and E.S.S. analyzed the Summit2010 and 20D ice core chemistry, respectively. Data analysis and its interpretation was performed by M.B.O., who wrote the manuscript with input from S.B.D. and L.D.T. All authors read and commented on the manuscript.

Competing interests

The authors declare no competing interests.

Figure legends

Figure 1: Strong covariability between Greenlandic [MSA] records (a) The twelve individual [MSA] time series, plotted from most southerly- (bottom; 20D) to most northerly-situated (top; NGT-B21). All series are standardized relative to A.D. 1821-1985 (period of common overlap; *z*-units) with linear [MSA] trends computed for the overlapping A.D. 1821-1985 period. Shaded red envelopes denote the regressions' 90% confidence intervals during the period of common overlap, while dotted lines show extension of the regressions beyond this period. Note the site identification ("ID") numbers to the right of the time series. (b) Communality scores for the 12 sites' HYSPLIT-derived "airmass transport density" maps (Methods; see also Fig. 2 and Extended Data Figure 4) following factor analysis with varimax rotation (Supplementary). Sites are grouped by whether their incoming marine airmasses are derived predominantly from the Irminger (factor #1; x-axis) or Labrador Seas (factor #2; y-axis). Locations of the 12 sites on the GrIS are provided in the inset, with each site-ID color coded with respect to its factor #1 communality score (i.e., Irminger Sea relative influence). Ice core sites influenced predominantly by airmasses of Irminger Sea origin are denoted using red hues ($n = 7$), while the predominantly Labrador Sea-influenced sites are denoted using blue hues ($n = 5$; see also Extended Data Figure 6).

Figure 2: [MSA]-PC1 and atmospheric back-trajectory modeling of probable MSA source regions a) Top panel: Time series of [MSA]-PC1 (black (red) line at 1-yr (10-yr lowpass-filtered) resolution with bootstrap-based 95% confidence interval; $n = 10{,}000$; Methods). Units (z) denote standard-variance. Bottom panel: Normalized probability histogram illustrating the variance explained by [MSA]-PC1 following 10,000 bootstrap-sampling principal component-tests (red; Methods). Also shown is the null-distribution of PC1-explained variance (grey) following 10,000 PCA tests conducted upon pseudo-random surrogate [MSA] datasets, revealing the [MSA]-PC1 series to be significantly different from noise at the $p = 0.002$ level (one-sided two-sample Kolmogorov-Smirnov test). The inset map shows each site's position on the GrIS and its homogenous correlation with [MSA]-PC1 over the period A.D. 1821-1985 (see Extended Data Table 1 for values). b) Site-weighted [MSA]-PC1 JJA marine-airmass transport density map (representing the relative probability of an oceanic airmass passing through a given atmospheric column *en route* to the GrIS; Methods), normalized on a 0-1 (least to most probable) scale. c) Site-weighted [MSA]-PC1 median atmospheric altitude for all ocean-situated JJA hourly trajectory locations over the period AD 1948-2013. The primary source of GrIS-[MSA] is assumed to overlap with regions representing high (low) airmass transport densities (atmospheric elevations; see also Extended Data Figures 4 and 5).

Figure 3: Strong agreement between subarctic Atlantic NPP, [DMS$_{SW}$], and [MSA]-PC1 (a) North Atlantic mean (log-transformed) satellite JJA VGPM-NPP rates (Methods; display data smoothed using a 3°x3° boxcar filter). Black contours show JJA [DMS$_{SW}$]-isopleths (nM) reproduced from ref. 23. Right panel denotes North Atlantic zonal NPP and [DMS$_{SW}$] averages. (b) Weighted (green; $n = 184$ degrees of freedom) and ordinary (grey; $n = 222$ degrees of freedom; Methods) least squares regression analysis of subarctic Atlantic [DMS$_{SW}$] vs. NPP rate; both regressions are highly significant ($p < 0.0001$), assuming a two-tailed Student's t-distribution with a t-statistic representing n-2 degrees of freedom. Regression values (r) represent Pearson product-moment coefficients. Shaded bands show the 95% confidence interval of the regression. Green circle diameter represents the relative weighting attributed to [DMS$_{SW}$] (Methods). (c) Leading EOF (20% of variance explained) of subarctic Atlantic summertime-integrated VGPM-NPP yields (A.D. 1998-2017; 50-65°N, 60°-10°W), showing strong overlap with the 95th-percentile [MSA]-PC1 airmass transport density (black bold line; Fig. 2b) and 5th-percentile [MSA]-PC1 median trajectory altitude isopleths (black dotted-dashed line; see Fig. 2c). (d) PC1-based projection of summertime VGPM-NPP yields (green) alongside the subarctic Atlantic-integrated NPP yield time series (yellow), overlain by [MSA]-PC1 (with grey bootstrap-based 95% confidence interval); all time-series are smoothed using a 5-year running mean. Individual (5-year smoothed) [MSA] records overlapping the satellite era are shown for comparison as light-grey lines. The yellow box in panel-c indicates the area shown in panel-a.

Figure 4: Multi-century decline of subarctic Atlantic productivity. (a) *i.* Observed[24] and reconstructed[25] NAO index. *ii.* Subarctic Atlantic "warming hole" SST anomalies[30], mean-centered relative to A.D. 1870-2016 (Extended Data Fig. 9) overlain with an extended SPG-index[22] (relative scale; Supplementary). *iii.* Reconstructed[6] and observed[8] AMOC index, overlain with 5-year averaged subarctic Atlantic freshwater storage anomalies[28] (±2σ; anomalies relative to A.D. 1955). *iv.* MSA]-PC1 productivity index (this study) overlain by 5-year smoothed NPP-PC1 (relative scale; Fig. 3c). The "Onset" range shows the estimated industrial-era initiation (1816 ± 11 yrs; Methods) of declining productivity. *v.* North Atlantic [Chl-α] reconstruction[11] (limited to the period of annually-contiguous data-availability, A.D. 1944-2006). *vi.* Standardized (*z*-score units relative to A.D. 1958-2016) indices of CPR-based diatom, dinoflagellate, and coccolithophore abundance (Methods). All thin (bolded) lines shown at 1-yr (10-yr lowpass-filtered) resolution, unless otherwise noted. (b) Weighted least squares calibration of 5-year smoothed [MSA]-PC1 and NPP-PC1 ($n = 12$ years; $r^2 = 0.63$; $p = 0.07$ after adjusting for reduced degrees of freedom; Methods). The regression weights are the inverse standard deviation of [MSA]-PC1 values (see also Fig. 3d). Blue and red distributions show the range of industrial-era onset and satellite-era [MSA]-PC1 values following 10,000 bootstrap tests (both distributions are normalized to their respective modes, with bold (dashed) vertical lines denoting the 50th (2.5/97.5th) percentiles). The grey-shaded region shows the 95% confidence interval of the regression parameters. The corresponding 95% confidence ranges of industrial-era onset and satellite-era annual NPP-yields are projected as vertical bands to the right, suggesting a mean ~10 ± 7% NPP decline (±2σ) over the industrial-era.

Methods

[MSA] record collection, analysis, and preprocessing

Twelve methanesulfonic acid (MSA; CH_3SO_3H) concentration ([MSA]) ice core records were compiled from sites situated on the Greenland Ice Sheet (GrIS; $n = 12$ records). MSA is measured at trace concentrations in polar ice via its constituent anion, methanesulfonate[31-32] (MS^-; $CH_3SO_3^-$). The five previously published [MSA] records used in this compilation were measured using either conventional ion-chromatography (IC) techniques (20D, ref. 31; NGRIP, ref. 33; GRIP93a, ref. 18) or electrospray ionization with triple quadrupole mass spectrometry (ESI–MS–MS; Summit2010 and TUNU; ref. 34). Specific details on the measurement techniques can be found in the original studies (Extended Data Table 1). Six out of seven of the remaining (previously unpublished) [MSA] records were measured using IC. Measurement of MS^- in the GC record[16] was conducted at Wheaton College (MA, USA) with analytical and core-sampling procedures identical to those described in ref. 35. Records derived from the Northern Greenland Traverse (NGT-B16, -B18, -B20, -B21, and -B26) were analyzed at the Alfred-Wegener-Institute following the methodology of ref. 36. The remaining unpublished [MSA] record, D4, was also analyzed via ESI–MS–MS at the Desert Research Institute following ref. 34.

Records were selected with the criterion that the records must i) be of moderate to high temporal resolution (measured at ≤3 years sample^{-1}; note 10/12 records exist at ≤1 year resolution; Fig. 1a), ii) be well-dated (<5 year estimated uncertainty at the deepest portions of the records presented), and iii) represent >100 years of continuous length within the period A.D. 1767-2013. Aspects of all ice cores have been previously published, such that information on each ice core's dating methodology can be found within references listed in Extended Data Table 1. Prior to analysis of the [MSA] records, each was linearly interpolated to a resolution of one year. The period of common overlap for all 12 records is A.D. 1821 – 1985. It is assumed that dating uncertainties amongst records are approximately normally distributed, such that dating inconsistencies are effectively averaged out during dimensional reduction. Similarly, although interior Greenlandic ice core [MSA] records are known to experience post-depositional vertical migration, migration directionality is not systematic and may occur in either the (atmosphere-oriented) up or down direction depending primarily on local cationic soluble impurity concentration gradients; ref. 37). As such, we assume that potential migration-based "skewing" of the original [MSA]-signal is largely minimized during data reduction. Finally, while contentions of post-depositional volatile losses of MSA have been reported at high-acidity and low accumulation sites in Antarctica (nominally, <0.10 kg m^{-2} yr^{-1}; ref. 38), such losses are likely largely inhibited across the GrIS, where relatively high accumulation rates, as well as low acidity summertime layers, prevail (Extended Data Table 1).

Note that a decision was made not to analyze MSA fluxes. This was due primarily to the lack of high-resolution accumulation data for all 12 sites. However, recent century-scale reconstructions of GrIS accumulation rate, derived from both inland[39-40] and near-coastal[41] ice cores, do not generally support evidence for spatiotemporally-synchronous shifts in accumulation across Greenland, nor to our knowledge regional multicentury accumulation increases necessary to promote (via dilution) the multi-century decreases in the [MSA] records presented here. Similarly, although MSA is highly hygroscopic and thus generally believed to be primarily wet-deposited on the GrIS[31], due to the lack of high-resolution accumulation data we are inhibited from quantitatively discerning the precise partitioning of [MSA] between wet and dry deposition at most (11/12, void 20D) sites. Nonetheless, our necessary assumption of negligible long-term changes in MSA depositional partitioning when spatially-averaged across the GrIS appears valid, given in particular the strong temporal covariation in [MSA] across differing GrIS moisture source regions (Extended Data Figure 6; Extended Data Table 1).

Extraction of [MSA]-PC1 and uncertainty estimation

We used an Empirical Orthogonal Function- (EOF) based data infilling routine[42] to infill missing values in the [MSA] records prior to signal extraction. In our study, missing values occur at the extremities of the records, and thus represent records that either i) were collected prior to A.D. 2013, or ii) did not extend as deep as A.D. 1767. Under the criterion that the oldest PC1 age (i.e., A.D. 1767) represents the oldest age where >75% of Greenlandic [MSA] records remain, <8% of data points amongst the 12 records required infilling. For the more recent portion of the PC1 series, we relaxed our 75% record-retention criterion to enable greater temporal-overlap with satellite observations (c. 1998). While this relaxation did, in general, invoke a trade-off with declining precision in the [MSA] signal extraction for satellite-interval years (as encapsulated by slightly enlarged [MSA]-PC1 confidence interval widths; Fig.'s 2a and 3d), we nonetheless expect our PC1 extraction to be robust given the strong satellite-era coverage of the Summit2010 record, the largest variance-contributor to [MSA]-PC1 (Table 1).

The EOF data infilling procedure[42] accounts for covariability between, as well as autocovariance within, individual MSA records, such that strong covariability between two records during a period of common overlap should result in imputed values of comparable covariance between the two records, should one of the records require infilling during a time period where data exists in the other. By such, the autocovariance structure of imputed values within that record should jointly reflect the autocovariance of that record's measured (that is, non-missing) values. The data infilling procedure[42] was conducted as follows: all records were standardized to unit variance and centered to mean zero over their period of common overlap, 1821 – 1985 A.D.

Missing values were set to zero (an unbiased *a priori* value), and the resultant matrix decomposed into left (temporal EOFs) and right (spatial EOFs) singular vectors using the method of Singular Value Decomposition (SVD). The missing (zero) values were then recovered by replacing the zero values with infilled values of the reconstructed [MSA] data matrix, following truncation of both EOF vector spaces. The number of EOFs retained for data infilling was obtained using a Monte Carlo cross-validation approach, whereby 5% of the [MSA] data points were withheld at random and iteratively reconstructed with a progressively less truncated EOF vector space until a specified convergence criterion was met (RMSE < 10^{-8}; ref. 42).

We tested the sensitivity of the cross-validation procedure across a large number of data infilling procedures using the [MSA] Greenlandic array, and found that EOF-based data infilling[42] routinely and robustly reproduced much of the low frequency variance of the [MSA] dataset across separate tests. However, slight variations in the magnitude of imputed values could occur between tests, an expected result due to the finite size of the [MSA] dataset used for cross validation. More specifically, variations in the optimal number of EOFs retained for the imputation of missing values could lead to small differences in the fraction of the original variance restored in the imputed [MSA] values between tests. In our case, the number of retained EOF's varied most often between 2 – 4, representing ~50-65% of the [MSA] variance. Since the amount of variance restored back into the imputed data will always be less than the original data, a method was required to restore remaining variance. To do so, we adopted an approach similar to ref. 15, whereby for each test we divided the infilled [MSA] data matrix into "signal" and "noise" components. The signal represents the "retained" [MSA] data matrix, constructed by applying the inverse EOF transform to the [MSA] dataset using only as many EOF's as was determined to optimally construct the imputed values. Conversely, the noise represents the "residual" [MSA] data matrix constructed by applying the inverse EOF transform with the remaining EOF's. We applied the method of Cholesky factorization to the noise component of each record, in order to produce pseudo-random noise vectors, i.e., randomized vectors with autocorrelation identical to each record's noise component, that could be added back to the imputed values in sequence and restore variance to the solution. In practice, our pseudo-random variance-restoration routine encourages enlarged uncertainty attribution in portions of the [MSA]-PC1 record requiring data-infilling (i.e., its extremities; see Figures 2a and 3d).

We incorporated a probabilistic principal component analysis in order to reduce "noise" amongst the 12 [MSA] records and better extract a meaningful mode of common variability, as well as to provide insight into the spatial distribution of homogenous [MSA] signals across the GrIS (e.g., Fig. 1c; Extended Data Figure 3; Supplementary). Extraction of the Greenlandic [MSA]-PC1 signal, including estimation of its confidence intervals, was conducted via the following procedure: i) The [MSA] dataset, $\boldsymbol{X_i}$, was centered to mean zero and standardized to unit variance, with missing values in $\boldsymbol{X_i}$ set to 0. ii) Missing values in $\boldsymbol{X_i}$ were statistically infilled following ref. 42, with pseudo-random variance restoration in the imputed values enforced. iii) Step 2) was repeated for an additional 99 realizations ($n = 100$ realizations total), with each $\boldsymbol{X_i} = \boldsymbol{X_1}, \boldsymbol{X_2}, \ldots \boldsymbol{X_n}$ stored for later use. iv) For each $i = 1, 2, \ldots n$ imputed [MSA] datasets, $j = 1, 2, \ldots n$ surrogate [MSA] datasets of equal dimension were created using uniform-random sampling with replacement of the [MSA] records (i.e., a "bootstrap" approach; ref. 43). v) PCA was performed on each $\boldsymbol{X_{i,j}}$ ($n^2 = 10^4$) surrogate [MSA] dataset, transformed using orthogonal Procrustes rotation in order to correct for (Eigen-transform) rotational ambiguity[43], and the PC1 extracted and stored. vi) The confidence intervals were computed using the 2.5th – 97.5th percentiles of the PC1 distribution (representing all n^2 tests). The "best-fit" PC1 signal represents the median fit (50th percentile) of the distribution. For a comparison with alternate methods of missing data estimation and [MSA]-PC1 extraction, the reader is referred to the Supplementary.

Attribution of probable MSA source regions

Changes in atmospheric circulation and windiness can affect aerosol transport and deposition across the GrIS, impacting the fidelity of ice core climate records across various timescales[32-34,44-49]. Heterogeneous signals existing across our 12 GrIS-[MSA] records, due to localized productivity and (or) atmospheric variations that are particularly salient across interannual to subdecadal timescales[44-46], are largely suppressed by dimensional reduction of the 12 [MSA] records into [MSA]-PC1 (Extended Data Fig. 3) and through multiyear smoothing of the PC1-series thereafter (i.e., 5-year smoothing; Fig. 2d; Extended Data Fig. 7).

Over longer, multidecadal to centennial timescales, current evidence[25-26] does not generally support significant shifts in recent internally-driven, regional lower-atmospheric dynamics, suggesting the primary emission source should underlie the most probable ("mean state") trajectory pathway taken by low-lying Greenland-bound air parcels when integrated across several decades (see also Supplementary 1c). To support this latter suggestion, we used the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory's Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, version 4.9 (ref. 50) to enable estimates of probable marine source regions of MSA at each site. HYSPLIT employs a joint Lagrangian-Eulerian approach, in which numerical singularities, or atmospheric "particles", are subjected to a time-variant, spatially fixed 3-dimensional gridded wind field across a time-invariant land-surface field, and tracked backwards in time at hourly-time steps.

Particle trajectories were forced atmospheric wind data from the National Centers for Environmental Protection and Atmospheric Research (NCEP/NCAR) global atmospheric reanalysis dataset[51], gridded at 2.5˚x2.5˚ resolution over 17 pressure levels (1000, 925, 850, 700, 600, 500, 400, 300, 250, 200, 150, 100, 70, 50, 30, 20, and 10 hPa). The HYSPLIT model forced using the NCEP/NCAR wind-reanalysis has been shown to provide comparable results to HYSPLIT trajectories forced using higher-resolution (e.g., 1˚-gridded) wind reanalysis products from the European Centre for Medium-Range Weather Forecasts (ref. 44).

Since position errors of an individual air mass back-trajectory are estimated to upwards of 30% of distance travelled[45-46], a probabilistic approach was taken here, in which a large-number of trajectories were computed and integrated into "airmass transport density maps". In order to focus on low elevation air masses (~0–1000 m), which are assumed to be more representative of regional marine-derived moisture and aerosol sources, all particle back-trajectories were initialized from a height of 500 meters above ground level and released daily during the months June-July-August from A.D. 1948-2013. In total, 6,121 trajectories were released above each ice core site (i.e., 73,452 total), and tracked hourly for 7 days prior to the particle release date, coinciding with the approximate atmospheric lifetime of MSA[46]. At the end of each model trajectory simulation, all hourly trajectory locations situated over ocean were summed in discrete 1˚x1˚ bins and area-normalized to produce the marine-airmass transport density grids, nominally representing the relative probability that any given trajectory endpoint would be situated over a given grid-cell at any point along a trajectory. Due to the inherent concentric partitioning of trajectory end-points around the trajectory release point, each grid-cell within the transport density grid was then normalized by its inverse radial distance from the trajectory release point to remove its central tendency[49]. Finally, all airmass transport density grids were normalized on a 0 – 1 (least to most probable) relative scale. In addition, the median particle trajectory height for each grid box was computed, in order to target regions consisting of predominantly low-lying oceanic airmasses, and thus those originating within the marine boundary layer.

In order to achieve an airmass transport density (and median atmospheric elevation) grid statistically representative of the [MSA]-PC1 series, we composited the 12 sites airmass transport density grids (and median atmospheric elevation grids) into a single map. This was achieved by weighting the 12 individual airmass transport density grids (and median atmospheric elevation grids) by each site's squared correlation (i.e., fraction of variance shared) with the [MSA]-PC1 signal over the common-overlap period A.D. 1821-1985 (Extended Data Table 2) prior to compositing. Note that the inferred source region, situated over the central-NE subarctic Atlantic in the vicinity of the Irminger and Icelandic Basins (Fig. 2c-d), is also considerably removed from the summertime sea ice marginal front where GrIS-deposited MSA origination has previously been attributed (ref. 34). For a more in-depth analysis of the possible maritime source regions of Greenlandic MSA on a per-site basis, the reader is referred to the Supplementary (see also Extended Data Figure 5).

Correlation analysis of subarctic Atlantic [DMS_{SW}] measurements to satellite-NPP

We compared surface seawater DMS concentrations ([DMS_{SW}]) to satellite derived net primary productivity (NPP) estimates (Fig. 2b) using measurements of [DMS_{SW}] compiled within the NOAA Global Surface Seawater DMS Database[23,52] for the period Jan 1 1998 to Dec 31 2016. This period was chosen in order to overlap with ocean color measurements from leading satellite sensors (e.g., SeaWIFS: late 1997 – 2009, and MODIS-AQUA: mid 2002 – present; Supplementary). It is important to note that no quality control on the [DMS_{SW}] measurements compiled in the database currently exist, due to the lack of [DMS_{SW}] measurement protocols, or inter-calibration methodologies[23]. Rather, in order to remove anomalous [DMS_{SW}] values, measurements representing the middle 95% of concentrations were retained for analysis. This resulted in 30,047 measurements. As [$\mathrm{DMS_{SW}}$] values archived within the database are most often clustered in space and time, values were binned monthly at 1˚x1˚ gridded resolution, log transformed to achieve normality and heteroscedasticity, and averaged. This procedure resulted in a reduction from 30,047 global measurements to 3045 unique global data points, a much smaller subset of which (n = 224) derives from the subarctic Atlantic (50-65˚N; 60-10˚W). We then upscaled (via 2-dimensional linear interpolation) the 1/6˚x1/6˚ gridded estimates of log-transformed ocean Net Primary Productivity (NPP), taken from the popular Vertically Generalized Production Model (VGPM; ref. 12), onto a centered 1˚x1˚ spatial grid. All unique [$\mathrm{DMS_{SW}}$] data points were regressed against the corresponding (log-transformed and standardized) NPP 1˚x1˚x1-month grid point using ordinary least squares. For [$\mathrm{DMS_{SW}}$] grid points in the subarctic Atlantic with more than one observation (n = 186 out of 224 total measurements), linear regression analysis was conducted via weighted least squares (WLS; where weights represent the inverse standard error of each average [$\mathrm{DMS_{SW}}$] value measured within a given 1˚x1˚x1-month bin) against the corresponding NPP value. An analysis of the relationship between global vs. subarctic Atlantic [$\mathrm{DMS_{SW}}$] and NPP can be found in Extended Data Figure 6 (see also the Supplementary).

Processing of Continuous Phytoplankton Recorder (CPR) survey data

For inferences of subarctic Atlantic phytoplankton abundance changes occurring since the mid-20th century, we use data from the Continuous Phytoplankton Recorder (CPR) survey. As reviewed by ref. 53, the CPR remains the most extensive (in spatial scale, taxonomic scope, and time period covered) independent ocean-biological monitoring program in current existence, having recorded the abundance of nearly 700 unique taxa since A.D. 1931. The sampling methodology consists of towing a filtering device at ~10 m depth along standard shipping routes using ships of opportunity, where each sample corresponds to 10 nautical miles (~18 km), or ~3$\mathrm{m^3}$ of filtered water.

Phytoplankton are collected on a 270 μm mesh, a size originally chosen to provide broad representation of planktonic species, including both larger predatorial functional groups (e.g., copepods, pteropods, and small crustaceans) and large-diameter autotrophic phytoplankton. Despite this mesh size, smaller planktonic species – including coccolithophores (~10 μm) and diatoms (~10-200 μm) – are also consistently captured on the silk mesh and recorded for abundance. Importantly, because the sampling methodology has remained relatively unchanged since the survey's inception, consistency of planktonic time series has been correspondingly maintained, and relative changes in planktonic abundance are considered to be generally robust irrespective of size and (or) functional group[53]. On the other hand, given the host of complicating factors pertaining to the collection and counting of different sized microorganisms in a given measurement (e.g., planktonic active avoidance or escape, mesh-clogging, cell visibility; see ref. 53), as well as the associated challenge therein of converting relative abundance measurements to absolute abundance, CPR measurements must nonetheless be cautioned as semi-quantitative by nature.

Here, we assess CPR products of monthly total diatom (1958-2016), dinoflagellate (1958-2016), and coccolithophore (1993-2016) abundance within preexisting CPR standard regions situated over the subarctic Atlantic (i.e., 50-65˚N, 60-10˚W, 14/41 CPR standard regions: A6, A8, B5-8, C5-8, D5-8; see Extended Data Fig. 3 or ref. 53). We targeted coccolithophore, dinoflagellate, as well as diatom relative abundances as these functional groups share both a known association to DMS production[19] and, collectively, are believed to comprise the bulk-abundance of autotrophic biomass in the subarctic Atlantic regions (e.g., ref.'s 17, 53). Towards this latter point, the decision to analyze each group was also of pragmatic intent, with each providing adequate CPR spatiotemporal coverage in most subarctic Atlantic regions over recent decades[53]

(Extended Data Figure 3). Conversely, since larger-diameter heterotrophs such as copepods and other zooplankton are not directly linked to DMS production (voiding their indirect association via sloppy-grazing and excretion[19]), and furthermore raise additional issues of systematic sampling bias due to, e.g., CPR inlet active-avoidance and escape[53], we did not directly consider these higher-order functional groups within our assessments (see also ref. 17).

As noted in ref. 53, a potential bias in decadal time-series of CPR data arises from the gradual increase in Atlantic shipping speeds since the mid-20th century. This shipping speed increase is believed to have had a systematic, and near-linear, negative effect on the amount of water filtered through CPR devices, thereby (negatively) biasing long term relative abundance trends[54]. As such, we correct for this potential bias using conservative (i.e., extreme case) empirical relationships established by ref. 54 between increasing mean northeastern-Atlantic shipping speed trends (0.09 knots year^{-1} since 1958) and volume water filtered ($-0.26\ \text{m}^3\ \text{knot}^{-1}$; all corrections made relative a mean filtered-water volume of $3.16\ \text{m}^3$ in AD 1990). As shown in Extended Data Figures 3c-d, this ship-speed bias adjustment imparts only minor adjustments on the “raw” CPR abundance data over the time period considered.

WLS regressions of CPR abundance against [DMS_{SW}] (Extended Data Fig. 6) were conducted following the procedure described for [DMS_{SW}] vs. NPP (above), the primary difference being that monthly [DMS_{SW}] values were instead averaged within entire CPR standard regions (as opposed to degree latitude-longitude bins), prior to regression. Relationships between [DMS_{SW}] and CPR-abundance were not found to be significantly different when using either the (ship-speed) bias-adjusted or raw CPR data (Extended Data Figure 3).

Time series of annual subarctic Atlantic phytoplankton abundance (shown in Fig. 4a) were estimated by first calculating annual means in each CPR standard region containing ≥8 months of data (Supplementary). We report CPR time series as the simple area weighted average of each standard regions' CPR abundance data across the subarctic Atlantic (which vary substantially in size). A comparison of subarctic Atlantic CPR time series – for both summertime- and annual-based measurements – to alternative probabilistic and deterministic data infilling and compositing techniques that better-account for regional-sampling biases are provided in Extended Data Fig. 3 (see also the Supplementary for an extended discussion).

Time series statistical significance testing

Statistical significance levels for all reported time series correlations (Pearson's r in all instances) were computed using the nonparametric Monte Carlo-based method of ref. 55, unless noted otherwise. We created $N = 10,000$ pseudo-random surrogate series of the first series by computing its Fourier transform, randomly varying the phase of its Fourier modes between 0 and 2π, and then computing the inverse transform, thereby retaining the exact autocorrelative properties (i.e., power spectrum preservation) of the original series. Statistical significance was then estimated by computing N psuedo-random correlations with the original second series, and by calculating the exceedance probability (i.e., inverse percentile) of achieving a correlation-magnitude greater than the original by chance alone. Note that the maximum degree of significance that can be achieved using this method is $p < N^{-1}$, i.e., $p < 0.0001$ represents an observed correlation-magnitude greater than all N pseudo-random correlations (e.g., Extended Data Table 1).

Productivity decline onset timing

The onset timing of the industrial-era productivity decline was estimated using the SiZer (SIgnificant ZERo crossings of derivatives) methodology[56], conducted in a manner similar to that described in ref.'s 14 and 16. Namely, we calculated the median significant ($p < 0.1$) onset of sustained (i.e., requiring the sign of the trend to persist into present) [MSA]-PC1 decline following pre-filtering of the series across a range of Gaussian kernel filters. We assessed 26 filters incrementally distributed from 15-40 year bandwidths. To alleviate edge-effect biases stemming from our comparably short[14,16] time series, we mandated each productivity-decline onset age to be at least one filter-width greater (i.e., more recent than) the oldest age of our time series (i.e., A.D. 1767). As such, our estimated industrial-era productivity-decline onset, A.D. 1816 ± 11 years (±2 median absolute deviations), represents the SiZer solution using a smaller subset (14/26) of the originally-filtered [MSA]-PC1 series.

Methods References

31. Whung, P. Y., Saltzman, E. S., Spencer, M. J., Mayewski, P. A. & Gundestrup, N. Two-hundred-year record of biogenic sulfur in a south Greenland ice core (20D). *J. Geophys. Res.* **99,** 1147–1156 (1994).
32. Saltzman, E. S., Whung, P.-Y. & Mayewski, P. A. Methanesulfonate in the Greenland Ice Sheet Project 2 Ice Core. *J. Geophys. Res. Ocean.* **102,** 26649–26657 (1997).
33. Andersen, K. K. *et al.* High-resolution record of Northern Hemisphere climate extending into the last interglacial period. *Nature* **431,** 147–151 (2004).
34. Maselli, O. J. *et al.* Sea ice and pollution-modulated changes in Greenland ice core methanesulfonate and bromine. *Clim. Past* **13,** 39–59 (2017).
35. Criscitiello, A. S. *et al.* Ice sheet record of recent sea-ice behavior and polynya variability in the Amundsen Sea, West Antarctica. *J. Geophys. Res. Ocean.* **118,** 118–130 (2013).
36. Fischer, H., Wagenbach, D., Kipfstuhl, J. Sulfate and nitrate firn concentrations on the Greenland ice sheet: 1. Large-scale geographical deposition changes. *J Geophys Res.* **103**, 21927-21934 (1998).
37. Osman, M., Das, S. B., Marchal, O. & Evans, M. J. Methanesulfonic acid (MSA) migration in polar ice: Data synthesis and theory. *Cryosphere* **11,** 2439–2462 (2017).
38. Weller, R., F. *et al*. Postdepositional losses of methane sulfonate, nitrate, and chloride at the European Project for Ice Coring in Antarctica deep-drilling site in Dronning Maud Land, Antarctica, *J. Geophys. Res.*, **109**, D07301 (2004).
39. Mernild, S. H. *et al.* Greenland precipitation trends in a long-term instrumental climate context (1890-2012): Evaluation of coastal and ice core records. *Int. J. Climatol.* **320,** 303–320 (2014).
40. Andersen, K. K. *et al.* Retrieving a common accumulation record from Greenland ice cores for the past 1800 years. *J. Geophys. Res. Atmos.* **111,** (2006).
41. Mosley-Thompson, E., Readinger, C. R., Craigmile, P., Thompson, L. G. & Calder, C. a. Regional sensitivity of Greenland precipitation to NAO variability. *Geophys. Res. Lett.* **32,** 1–4 (2005).
42. Beckers, J. M. & Rixen, M. EOF calculations and data filling from incomplete oceanographic datasets. *J. Atmos. Ocean. Technol.* **20,** 1839–1856 (2003).
43. Babamoradi, H., van den Berg, F. & Rinnan, Å. Bootstrap based confidence limits in principal component analysis — A case study. *Chemom. Intell. Lab. Syst.* **120,** 97–105 (2013).
44. Sinclair, K.E., Bertler, N.A., Trompetter, W.J., and Baisden, W.T. Seasonality of Airmass Pathways to Coastal Antarctica: Ramifications for Interpreting High-Resolution Ice Core Records. *J. Climate,* **26**, 2065–2076 (2013).
45. Schlosser, E., Oerter, H., Masson-Delmotte, V. & Reijmer, C. Atmospheric influence on the deuterium excess signal in polar firn: Implication for ice-core interpretation. *J. Glaciol.* **54,** 117–124 (2008).
46. Scarchilli, C., Frezzotti, M. & Ruti, P. M. Snow precipitation at four ice core sites in East Antarctica: Provenance, seasonality and blocking factors. *Clim. Dyn.* **37,** 2107–2125 (2011).
47. Sharma, S. *et al.* Influence of transport and ocean ice extent on biogenic aerosol sulfur in the Arctic atmosphere. **117,** (2012).
48. Hezel, P. J. *et al.* Modeled methanesulfonic acid (MSA) deposition in Antarctica and its relationship to sea ice. *J. Geophys. Res. Atmos.* **116,** 1–18 (2011).
49. Criscitiello, A. S. *et al.* Marine aerosol source regions to prince of Wales Icefield, Ellesmere Island, and influence from the tropical Pacific, 1979-2001. *J. Geophys. Res.* **121,** 9492–9507 (2016).
50. Stein, A. F. *et al.* NOAA's HYSPLIT atmospheric transport and dispersion modeling system. *Bull. Am. Meteorol. Soc.* **96,** 2059–2077 (2015).
51. Kalnay, E. *et al.* The NCEP/NCAR 40-Year Reanalysis Project. *Bull. Am. Meteorol. Soc.* **77,** 437–471 (1996).
52. Kettle, A. J. *et al.* A global database of sea surface dimethylsulfide (DMS) measurements and a procedure to predict sea surface DMS as a function of latitude, longitude, and month. *Global Biogeochem. Cycles* **13,** 399–444 (1999).
53. Richardson, A. J. *et al.* Using continuous plankton recorder data. *Prog. Oceanogr.* **68,** 27–74 (2006).

54. Jonas T. D., *et al*. The volume of water filtered by a Continuous Plankton Recorder sample: the effect of ship speed, *J. of Phytoplankton Research* **26**, 1499–1506 (2004).
55. Ebisuzaki, W. A method to estimate the statistical significance of a correlation when the data are serially correlated. *J. Clim.* **10,** 2147–2153 (1997).
56. Hannig, J. & Marron, J. S. Advanced distribution theory for SiZer. *J. Am. Stat. Assoc.* 101, 484–499 (2006).
57. Westberry, T., Behrenfeld, M. J., Siegel, D. A. & Boss, E. Carbon-based primary productivity modeling with vertically resolved photoacclimation. *Global Biogeochem. Cycles* **22,** 1–18 (2008).
58. Rayner, N. A., Parker, D. E., Horton, E. B., Folland, C. K., Alexander, L. V., Powell, D. P. Global analyses of sea surface temperature, sea ice, and night marine air temperature since the late nineteenth century. *J. Geophys. Res.* **108,** 4407 (2003).
59. Josey, S. A. *et al.* The Recent Atlantic Cold Anomaly: Causes, Consequences, and Related Phenomena. *Ann. Rev. Mar. Sci.* **10,** 475–501 (2018).
60. McConnell, J. R. *et al.* 20th-Century Industrial Black Carbon Emissions Altered Arctic Climate Forcing. *Science* **317,** 1381–1384 (2007).
61. Weißbach, S. *et al.* Spatial and temporal oxygen isotope variability in northern Greenland – implications for a new climate record over the past millennium. *Clim. Past* **12,** 171–188 (2016).

Data and code availability

Annual ice core [MSA] data used in this study are available via the NSF Arctic Data Center (http://arcticdata.io). Additionally, source data for Figures 1, 2, and 4 are available in the online version of this paper. Code used for [MSA] signal extraction, Monte Carlo correlation analysis, HYSPLIT analysis, and CPR analysis is available from M.B.O. upon request. Code for SiZer change-point analysis was modified after ref. 4 (https://www.nature.com/articles/nature19082) and is available from M.B.O. upon request. Availability of CPR plankton-abundance data is made possible by the Sir Alistar Hardy Foundation for Ocean Science (https://www.cprsurvey.org/; doi:10.7487/2018.29.1.1109 and doi:10.7487/2018.53.1.1118). Ocean productivity data are publicly available from https://www.science.oregonstate.edu/ocean.productivity/. Ocean [DMS_{SW}] data can be accessed via https://saga.pmel.noaa.gov/dms/. The HYSPLIT source-code and NCAR-NCEP reanalysis data can be downloaded at https://www.ready.noaa.gov/HYSPLIT.php.

Extended data legends

Extended Data Figure 1: Comparison of net primary productivity products. (a) Monthly integrated NPP (g C) across the subarctic Atlantic (50-65˚N, 60-10˚W, region highlighted as the yellow boxed region in a-d) for the SeaWiFS-VGPM and MODIS-VGPM NPP products (ref. 12; as shown in the main text), as well as independently-derived SeaWiFS- Carbon-based Productivity Model (i.e., CbPM; ref. 57; Supplementary) and MODIS-CbPM NPP products. (b) SeaWiFS and MODIS-derived NPP mean-seasonality (±2σ; $n = 20$ years; Jan. 1998- Dec. 2017) for the VGPM and (c) CbPM datasets. Note that four months – Nov-Dec-Jan-Feb – experience partial polar darkness over the subarctic Atlantic latitude bands (50-65˚N), leading to systematic underestimates of productivity during these months. (d) VGPM and (e) CbPM based linear-regressions with ship-based [DMS_{SW}] measurements (reminiscent of Fig. 3b of the main text; see Methods) using the MODIS- and SeaWiFS-NPP datasets. Shaded bands show the 95% confidence interval of the regression. Regression values (r^2) represent the squared Pearson product-moment coefficients.

Extended Data Figure 2: Seasonal representativeness of subarctic Atlantic VGPM-NPP satellite-era trends, and sensitivity to satellite sensor used. (a) Comparison of summertime-integrated (JJA) subarctic Atlantic VGPM-NPP yields for three different sensor estimates: a "SeaWiFS-dominant" estimate (red; 1998-2007 NPP estimates derived from the SeaWiFS sensor; 2008-2017 NPP estimates from the MODIS sensor), a MODIS-dominant estimate (blue; SeaWiFS-based data from 1998-2002, MODIS-based data from 2003-2017) and the composite stack (dark-grey; comprising the average of SeaWiFS- and MODIS-derived summertime VGPM-NPP estimates over their period of common annual overlap, 2003-2007). (b) Differential linear trend analysis of the composite summertime subarctic Atlantic NPP time-series from (a). No decadal-scale linear trends were found to be significant at $p < 0.05$, using a two-sided Student's t-test with n-2 degrees of freedom (where n represents the varying trend length in years). (c) EOF1 and (d) PC1 of summertime VGPM-NPP using the MODIS-dominant dataset, reminiscent of Fig. 2c-d from the main text. (e-f) as in (c) and (d), but showing EOF-results from the SeaWiFS-dominant summertime VGPM-NPP dataset. (g-l) As in (a-h), but showing annually-integrated VGPM-NPP estimates. All regression values (r) represent Pearson product-moment coefficients.

Extended Data Figure 3: Comparison of subarctic Atlantic CPR compositing techniques. (a-b) Data availability (A.D. 1958-2016) by CPR standard region (Methods) for (a) annually- (≥8/12 months/year of data required) and (b) summertime- (≥4/12 months/year of data during Apr-May-June-Jul-Aug-Sept) considered data. (c-d) Three approaches for compositing time series of CPR-based planktonic abundance, for both (c) annual- and (d) summertime-based data: AWA = Area Weighted Averaging; ISD = Inverse-Squared Distance-based data-infilling; EOF = Empirical Orthogonal Function-based data-infilling (ref. 42; see Supplementary). Thin dashed lines show raw annual relative abundance plankton concentrations, while bolded lines show "adjusted" relative abundances, to correct for potential long-term biases in the volume of water sampled by CPR devices[53-54]. Note that the annual AWA series is reproduced from Fig. 4a.

Extended Data Figure 4: HYSPLIT-computed summertime (JJA) airmass transport probability densities for each ice core site. Site-specific JJA marine-airmass transport density maps, representing the relative probability of an oceanic airmass passing through a given atmospheric column prior to its arrival at each site. All marine-airmass transport density maps are computed over the period A.D. Jan. 1st, 1948 to Dec. 31st, 2013 (i.e., 6121 JJA trajectories per site), and normalized on a 0-1 relative scale with 1 (0) indicating the most (least) probable airmass trajectory grid-point. Sites are shown counter-clockwise from most southerly- (20D; upper left) to most northerly- (NGT-B21; upper right) situated on the GrIS.

Extended Data Figure 5: HYSPLIT-computed summertime (JJA) airmass median elevation maps for each ice core site. Site-specific median atmospheric altitudes (meters above sea level) for all ocean-situated JJA hourly trajectory locations over the period AD 1948-2013. All maps were computed over the period A.D. Jan. 1st, 1948 to Dec. 31st, 2013 (i.e., 6121 JJA trajectories per site). Sites are shown counter-clockwise from most southerly- (20D; upper left) to most northerly- (NGT-B21; upper right) situated on the GrIS.

Extended Data Figure 6: Strong covariation between two [MSA] source-trajectory regions. Top panel: The twelve Greenland [MSA] records from Figure 1a annually-averaged across the two airmass-trajectory factor analysis-groupings from Figure 1b ($r = 0.63$, $p < 0.0001$; ref. 55); the r-value represents the Pearson product-moment coefficient. All records have been standardized (z-units) relative to their period of common overlap (A.D. 1821-1985). The shaded bands show ±1 standard error about the stack means. The grey line shows the composite (12-site) mean. Bottom panel: [MSA] record availability over time.

Extended Data Figure 7: Relation between [$\mathrm{DMS_{SW}}$], NPP, and CPR planktonic abundance. (a) Reproduced from Fig. 3b. (b) As in (a), but for globally regressed values. Diameter of circles represent the root of the relative weighting (inverse standard error of [$\mathrm{DMS_{SW}}$] measurement) used in the weighted least squares (WLS) regressions ($n_{WLS,subarctic} = 184$; $n_{WLS,global} = 2219$ degrees of freedom). Grey circles represent points used in the ordinary least squares (OLS) regression ($n_{OLS,subarctic} = 222$; $n_{OLS,global} = 3043$). (c) Probability density of global r values over $i = 1, 2, \ldots, 10{,}000$ degrees-of-freedom preserving (i.e., $n_i = 184$) bootstrap WLS regressions (Supplementary). (d-f) Linear regression analyses of subarctic Atlantic [$\mathrm{DMS_{SW}}$] vs. CPR-based (d) diatom, (e) dinoflagellate, and (f) coccolithophore abundance. In all regressions, the colored (grey) shaded region denotes the 95% confidence interval about the regression parameters for the WLS (OLS) regression. All WLS (OLS) regressions significant at $p < 0.005$ (<0.05), assuming a two-tailed Student's t-distribution with a t-statistic representing n-2 degrees of freedom. Regression values (r) represent Pearson product-moment coefficients.

Extended Data Figure 8: Industrial-era decline in subarctic Atlantic NPP and climatic influence. (a) Correlation matrix (Pearson product-moment coefficients, r) of planktonic and observed-climatic indices from Fig. 4a. Integers represent n, the years of overlap between paired series. Bold n-values represent significance at the 90% confidence level ($p < 0.1$; assuming a two-tailed Student's t-distribution with a t-statistic representing n-2 degrees of freedom). Bold n-values with an asterisk represent significance using a Monte Carlo-based Fourier phase-randomization procedure, a more stringent test to account for serial correlation (and hence varying degrees of freedom) amongst paired series (Methods). All 10-yr lowpass-filtered (bottom-left of diagonal), and linearly-detrended 10-yr lowpass-filtered (top-right of diagonal), series convolved using a Gaussian filter. Paired series with less than 20 years of overlap are denoted missing by "x". (b) WLS model of 5-year smoothed [MSA]-PC1 and summertime NPP-PC1 ($n = 12$ yrs; $r^2 = 0.72$; $p = 0.04$; significance estimated via the method of ref. 55, to adjust for the reduced degrees of freedom introduced from multiyear averaging). The regression weights are the inverse standard deviation of [MSA]-PC1 values. Histogram distributions denotes the range of industrial-era onset and satellite-era [MSA]-PC1 values following 10,000 bootstrap tests (distributions normalized to their maximum). The shaded band shows the 95% confidence interval of the WLS-regression parameters. The corresponding 95% confidence range of NPP rates (g C $\mathrm{JJA^{-1}}$) over the industrial-era onset and satellite-era are shown to the right. The analysis suggests an average ~14 ± 11% decline (±2σ) in summertime-integrated NPP yields since the industrial-era onset.

Extended Data Figure 9: Subarctic Atlantic sea-surface temperature (SST) analysis. Annual SST linear-trends in the (a) ERSST (v5; ref. 30) and (b) HadISST (v1.1; ref. 58) reanalyses. Grid points exhibiting a 147-year (A.D. 1870-2016) cooling trend within the subarctic Atlantic (50-65˚N; 60-10˚W; bolded blue outline) are outlined by black isopleths, and defined to encompass the Atlantic "Warming Hole" (ref. 59). (c) ERSST and HadISST anomalies (mean-centered relative to A.D. 1870-2016; $n = 147$ years) for the Northern Hemisphere (NH, top; ±1σ), the Atlantic Warming Hole (middle; ±1σ), and the difference between Warming Hole and Northern Hemisphere SSTs, representing the AMOC index as approximately defined in ref.'s 6 and 7 (bottom; ±1σ). Bolded AMOC time series are 10-yr (Butterworth) lowpass-filtered.

Extended Data Table 1: Geographical, physical, and glaciological information pertaining to each ice core [MSA] record. The third to last column provides the annual site-mean and standard deviation [MSA] values for the period of common overlap between records, A.D. 1821-1985 (n = 165 years). The second to last column provides the homogenous correlations (Pearson r) and significance level (p; ref. 55) between [MSA]-PC1 and the 12 GrIS-[MSA] records comprising it, computed over the period of common overlap. Note that correlation values are reproduced from color-coded values shown on the Figure 2a inset map.