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Key Points:
- We found regionally and temporally varying relationships between atmospheric DMS and air mass exposure to phytoplankton in the Arctic Ocean
- The DMS production capacity of the Greenland Sea was 3 times greater than that of the Barents Sea
- There is a higher abundance of DMS-producing phytoplankton in the Greenland Sea than in the Barents Sea during the phytoplankton bloom period

Supporting Information:
- Supporting Information S1
- Data Set S1

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Atmospheric DMS in the Arctic Ocean and Its Relation to Phytoplankton Biomass

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Abstract We recorded and analyzed the atmospheric dimethyl sulfide (DMS) mixing ratios at a remote Arctic location (Svalbard; 78.5°N, 11.8°E) during phytoplankton bloom periods in the years 2010, 2014, and 2015 and found varying regional relationships between the atmospheric DMS and the extent of exposure of the air mass to the phytoplankton biomass in the ocean surrounding the observation site. The DMS production capacity of the Greenland Sea was estimated to be a factor of 3 greater than that of the Barents Sea, whereas the phytoplankton biomass in the Barents Sea was more than twofold than that in the Greenland Sea. These apparently contradictory results may be induced by the occurrence of a greater abundance of DMS-producing phytoplankton in the Greenland Sea than in the Barents Sea during the phytoplankton bloom periods.

1. Introduction

Ocean biology may influence the Earth's climate through its effects on atmospheric composition due to the release of various organic compounds into the atmosphere (Gantt & Meskhidze, 2013; Meskhidze & Nenes, 2006; O'Dowd et al., 2004; Yoon & Brimblecombe, 2002). In particular, sulfur-containing aerosols provide an important source of cloud condensation nuclei in marine atmosphere in the polar region (Andreae et al., 1995; Chang et al., 2011; Chen et al., 2012; Ghahremaninezhad et al., 2016). The production of sulfate from the oxidation of dimethyl sulfide (DMS) was proposed as a negative feedback mechanism by which phytoplankton can modulate the properties of marine clouds (Charlson et al., 1987). DMS is mostly produced by marine phytoplankton and is the most abundant form of biogenic sulfur released from the ocean (Lovelock et al., 1972; Stefels et al., 2007). Global oceanic DMS emissions have been estimated to be 18–34 Tg S yr^{-1} (Lana et al., 2011), accounting for approximately 40% of the total sulfur flux (Simó, 2001). Specifically, the emission of oceanic DMS has a discernable impact on aerosol formation in the Arctic atmosphere during phytoplankton bloom periods (Park et al., 2017). As Arctic warming occurs in the future, the decrease in sea ice may alter the abundance and species composition of phytoplankton (Wassmann, 2011). These changes may cumulatively have cascading effects on oceanic DMS emissions and sulfur aerosol formation (Becagli et al., 2016; Dall' Osto et al., 2017; Gabric et al., 2017; Levasseur, 2013).

Regional associations between oceanic DMS emissions and phytoplankton biomass (expressed here as chlorophyll) can be useful in predicting oceanic DMS emissions from phytoplankton biomass data. However, an intrinsic hurdle to establishing such associations is that the production of the DMS precursor, dimethylsulfoniopropionate (DMSP), is highly species-specific (Keller et al., 1989; Park, Lee, Shin, Jeong, et al., 2014), and the conversion of DMSP to DMS is highly variable because this process occurs through complex processes (Kim et al., 2010; Park, Lee, Shin, Yang et al., 2014; Stefels et al., 2007). In practice, the atmospheric DMS mixing ratio can undergo abrupt transition between high and low values in response to rapid changes in the origin of the air mass reaching the site, or changes in meteorological conditions that determine the rate of emission of DMS from the ocean and its subsequent breakdown in the atmosphere through various chemical processes (Methven et al., 2001; Park et al., 2013). Together, these factors make it difficult to establish the unique relationship between atmospheric DMS and phytoplankton biomass.

The primary goal of the present study was to evaluate how the exposure of an air mass to phytoplankton biomass relates to the atmospheric DMS mixing ratios of the air mass reaching the Svalbard observation site, and

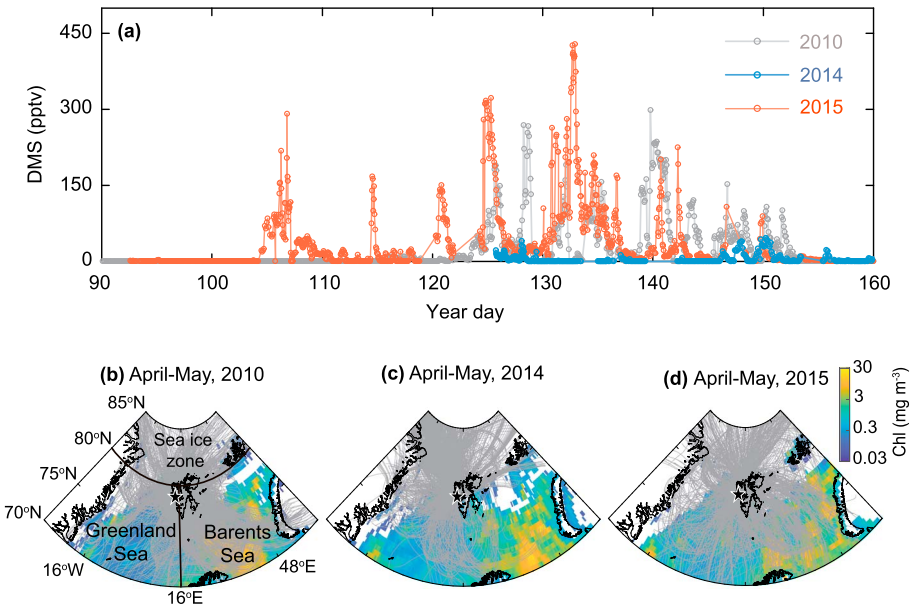

Figure 1. (a) The mixing ratios of atmospheric dimethyl sulfide (DMS) measured at the Zeppelin station in 2010, 2014, and 2015. (b–d) Monthly mean chlorophyll concentration during the month of April–May in 2010, 2014, and 2015, overlaid with the 2 day air mass back trajectories collected at hourly intervals. The star symbol indicates the location of the Zeppelin station (78.5°N, 11.8°E), Svalbard. The solid lines indicate the boundaries of the three subdomains: sea ice zone (north of 80°N), the Barents Sea (70°N–80°N, 16°E–50°E), and the Greenland Sea (70°N–80°N, 25°W–16°E).

to assess the relationships between the atmospheric DMS mixing ratio and the plankton biomass of the ocean region surrounding the Svalbard site during phytoplankton bloom periods. This analysis examined the atmospheric DMS mixing ratios determined continuously over a period of the phytoplankton bloom (April to May) in each of the 3 years (2010, 2014, and 2015). The biological exposure of the air mass reaching the observation site was estimated using meteorological parameters (air mass back trajectory, altitude, and speed) and satellite chlorophyll concentration data obtained from the ocean basins supposed to be sources for Svalbard DMS at the time of the atmospheric DMS measurements.

2. Materials and Methods

2.1. Data Sources (Atmospheric DMS, Chlorophyll, and Meteorological Parameters)

The atmospheric DMS analytical system was installed in March 2010 at an elevation of 474 m above sea level on Zeppelin Mountain, Svalbard (78.5°N, 11.8°E). The atmospheric DMS mixing ratio was measured at 1−2 h intervals from April to May in 2010, May in 2014, and April to May in 2015. The measurement periods approximately covered the phytoplankton bloom periods (e.g., Degerlund & Eilertsen, 2010; Sakshaug, 2004). The analytical system includes a component for DMS trapping and elution and a gas chromatography equipped with a pulsed flame photometric detector enabling DMS quantification (Jang et al., 2016). The detection limit of the analytical system was about 1.5 pptv in an air sample volume of ~6 L (Jang et al., 2016). The oceanic region adjacent to Svalbard was divided into three domains that included the sea ice zone (>80°N, low biological productivity), the Barents Sea (70°N–80°N, 16°E–50°E, epicontinental sea, high biological productivity), and the Greenland Sea (70°–80°N, 25°W–16°E, deep ocean basin, high biological productivity) (Figure 1).

The phytoplankton biomass of the three-ocean domains surrounding Svalbard was obtained by calculating the chlorophyll concentrations from the Level-3 product of Aqua Moderate Resolution Imaging Spectroradiometer at a 4 km resolution. The air mass back trajectories and meteorological parameters, including the pressure of the air mass and marine boundary layer, were obtained using the Hybrid Single-Particle Lagrangian Integrated Trajectories model (Draxler & Hess, 1998). The 2 day air mass back trajectories and hourly positions were determined and combined with satellite-based chlorophyll concentration data to show the 2 day evolution of the exposure of a given air mass to phytoplankton biomass.

2.2. Calculation of Air Mass Exposure to Chlorophyll

The DMS mixing ratio of an air mass reaching Svalbard depends primarily on two factors: (i) the area of ocean surface containing phytoplankton over which the air mass travels and (ii) the period of time during which the air mass is in contact with that area (Arnold et al., 2010; Park et al., 2013). The shorter period of contact (i.e., a greater air mass speed) means greater release of DMS and less oxidation of DMS in the atmosphere before it reaches the Svalbard site. In addition, a shorter duration of contact between the air mass and the phytoplankton biomass, in combination with a greater phytoplankton biomass per unit area, would result in a higher DMS mixing ratio. The "air mass exposure to chlorophyll" calculated here reflects the influences of phytoplankton biomass and atmospheric DMS oxidation on the DMS mixing ratio of the air mass reaching Svalbard. Note that we did not account for the effect of changes in the speed of the air mass because it is considerably smaller than the effect of DMS oxidation.

In calculating the air mass exposure to ocean chlorophyll, we assumed that satellite-based chlorophyll concentrations are a good proxy for phytoplankton biomass (Siegel et al., 2013) and that phytoplankton biomass was the major source of DMS to the marine atmosphere via enzymatic cleavage of phytoplanktonic DMSP to DMS (Simó, 2001). Therefore, the air mass exposure to ocean chlorophyll provides a good measure of the DMS production capacity of the ocean region through which air mass has passed. The air mass exposure to ocean chlorophyll (E_{chl}) at each time period over which we analyzed the atmospheric DMS mixing ratio was calculated using equation (1), below. The number of measured atmospheric DMS mixing ratios was 888 in 2010, 312 in 2014, and 941 in 2015.

$$E_{chl} = \frac{\sum_{t=1}^{48} Chl_t \cdot e^{-\alpha\left(\frac{t}{24}\right)}}{n} \quad (1)$$

Here Chl_t is the 8 day mean chlorophyll concentration within a radius of 25 km at a given time point (t = 1 to 48) along the air mass back trajectory, and n is the total number of time points for which valid chlorophyll values are available. We limited our analysis of air mass back trajectories to 2 days prior to the arrival of the air masses at the observation site because more than 60% of atmospheric DMS was reported to disappear within 2 days following airborne delivery due to photooxidation (Leck & Persson, 1996; Sharma et al., 1999). In a previous study the strongest correlation between air mass exposure to chlorophyll was found for a 2 day transport history and atmospheric trace gas mixing ratios including DMS (Arnold et al., 2010). The term $e^{-\alpha(t/24)}$ corresponds to the normalization of the photodecay, where α is the decay constant of DMS in the atmosphere due to photochemical processes. A value of 0.43 was used for α (35% DMS loss per day in Arctic atmosphere) (Leck & Persson, 1996).

For a given air mass back trajectory, the time points for which satellite-based chlorophyll concentration data were not available were excluded from the calculation, and the time points for which the pressure of the air mass was less than 850 hPa were assigned a zero chlorophyll value. A zero chlorophyll value was also assigned to the time points when the air mass passed over the Svalbard islands or regions covered by sea ice (Arnold et al., 2010).

2.3. Data Filtration

To avoid ambiguities in the relationship between the atmospheric DMS mixing ratio and the air mass exposure to biology, three sequential data filtration procedures were used to remove inappropriate data (Figures 2a–2e and S1). Any 2 day air mass back trajectory for which valid chlorophyll concentration data were not available for >50% of the time points were excluded from our analysis (first filtration). Among all 2 day air mass back trajectories that were not excluded by the first criterion, those having 2 day mean air mass speeds <3 m s^{-1} were also excluded (second filtration). The second filtration was applied because the low-speed air masses probably originated from coastal areas near Svalbard, or from fjords within Svalbard, where satellite-based chlorophyll data were either mostly unavailable or highly unreliable (Figure S4). Finally, all air mass back trajectories were grouped into one of three ocean domains by selecting only the 2 day air mass back trajectories that had >90% retention in a given ocean domain (third filtration).

Calculation of a given air mass exposure to chlorophyll may have been biased by exclusion of time points for which reliable chlorophyll values were not available. To minimize any undesirable effect of such data exclusion, we used monthly mean chlorophyll values if 8 day mean chlorophyll values were not available. As a

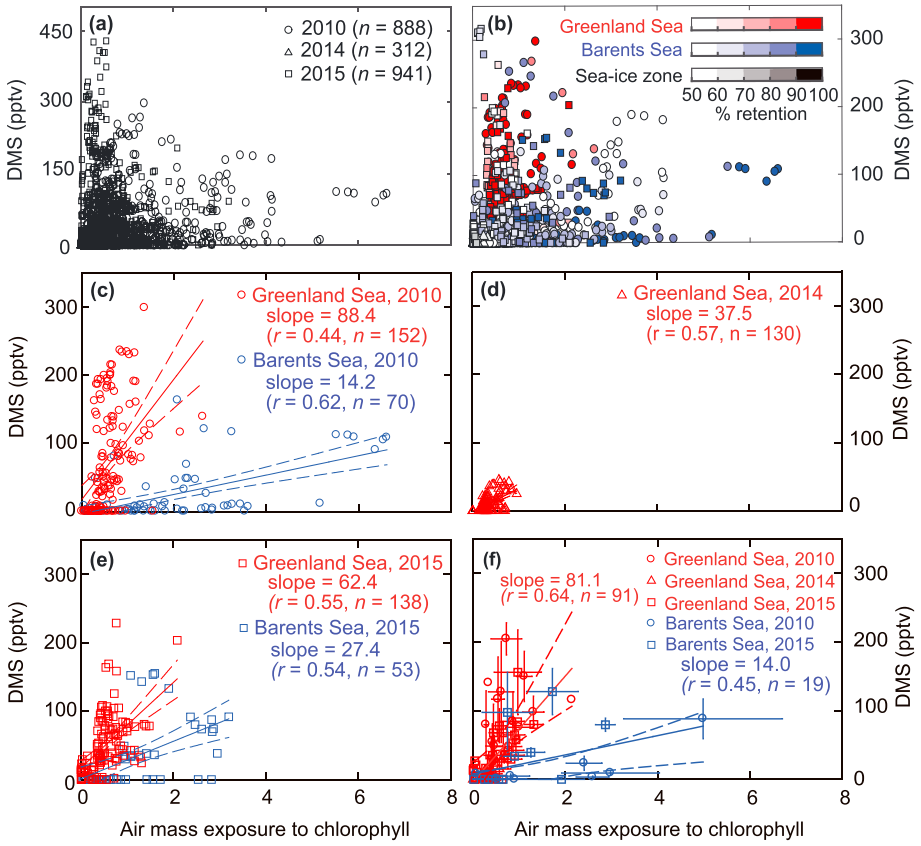

Figure 2. The mixing ratios for the atmospheric dimethyl sulfide (DMS) measured at the observation site as a function of the air mass exposure to chlorophyll. (a) All data sets obtained from April to May in 2010, 2014, and 2015. (b) All data shown in Figure 2a, excluding those corresponding to the 2 day air mass back trajectories for which valid chlorophyll data were not available for >50% of the time points (first filtration), and the 2 day air mass trajectories having mean air mass speeds <3 m s^{-1} (second filtration). The color bar indicates retention percentage (%) of air mass trajectories in a given ocean domain along the 2 day air mass back trajectory. (c–e) Data used for estimating the correlation between the atmospheric DMS mixing ratios and the air mass exposure to chlorophyll; this shows all data in (Figure 2b excluding data corresponding to air mass back trajectories that did not have >90% retention in a given ocean domain (third filtration). (f) The daily mean atmospheric DMS mixing ratio as a function of the air mass exposure to chlorophyll shown in Figures 2c–2e. Red symbols indicate the data set corresponding to the air mass that originated from the Greenland Sea. Blue symbols indicate the data set corresponding to the air mass that originated from the Barents Sea. The error bars indicate 1 standard deviation (1σ) from the daily mean values. The solid and dashed lines indicate the best fits and the 95% confidence intervals, respectively.

result, approximately 25% of the measured atmospheric DMS mixing ratios were used in the estimation of the correlation between the atmospheric DMS mixing ratios and the air mass exposure to chlorophyll (Figures 2 and S1). All of the 2 day air mass back trajectories used in the final estimation of the air mass exposure to chlorophyll for 2010, 2014, and 2015 are shown in Figure S5.

3. Results and Discussion

3.1. Atmospheric DMS Mixing Ratios at Svalbard

The atmospheric DMS mixing ratio observed at Svalbard showed striking differences from season to season and from year to year (Figure 1a). The DMS mixing ratios increased by 100–450 pptv in May 2010 and 2015, with considerable short-term (less than a few days) and long-term (1 week) variability; however, the DMS mixing ratios dropped rapidly by an order of magnitude and remained at that level thereafter. Throughout the winter, from September to April, the atmospheric DMS mixing ratio was nearly undetectable (not shown in Figure 1a, which was characterized by an analytical detection limit of 1.5 pptv DMS). Across all of the

3 year data sets, the atmospheric DMS mixing ratios varied on a time scale of <1 week, which could be explained in terms of the changes in the air mass origin, altitude, and speed (Park et al., 2013).

3.2. Phytoplankton in the North Atlantic Ocean Surrounding Svalbard

During the 3 years of observations (2010, 2014, and 2015), the phytoplankton biomass of the three ocean domains surrounding Svalbard differed considerably (Figures 1b–1d, S5, and S6). During the growing season between April and May, the phytoplankton biomass in the central Arctic Basin (sea ice zone) was the lowest (Figures 1b–1d), which was most likely due to short growing season as a result of extensive sea ice coverage. In contrast, the largest biomass was observed in the Barents Sea, followed by the Greenland Sea, which is a pattern previously reported by Sakshaug (2004). For example, the satellite-based chlorophyll concentration in the Barents Sea was >2 times the value found in the Greenland Sea (Figures 1b–1d and S5).

3.3. Relationship Between the Atmospheric DMS and the Air Mass Exposure to Chlorophyll

A positive correlation between the atmospheric DMS and the air mass exposure to chlorophyll was found during the phytoplankton bloom periods of the observation years, from April to May in 2010, $r = 0.44$ ($P < 0.05$, $n = 152$) for air masses originating from the Greenland Sea; $r = 0.62$ ($P < 0.05$, $n = 70$) for air masses originating from the Barents Sea (Figure 2c); in May of 2014, $r = 0.57$ ($P < 0.05$, $n = 130$) for air masses from Greenland Sea (Figure 2d); and from April to May of 2015, $r = 0.55$ ($P < 0.05$, $n = 138$) for air masses originating from the Greenland Sea and from April to May, $r = 0.54$ ($P < 0.05$, $n = 53$) for air masses originating from the Barents Sea (Figure 2e). More explicitly, the slope between the atmospheric DMS and the air mass exposure to chlorophyll approximately corresponded to the DMS production capacity of the ocean region. The slopes found in the Greenland Sea (88.4 for 2010, 37.5 for 2014, and 62.4 for 2015) were >3 times the values found in the Barents Sea (14.2 for 2010, not available for 2014, and 27.4 for 2015) (Figures 2c–2f).

These results indicate the occurrence of a greater abundance of DMS-producing phytoplankton in the Greenland Sea than in the Barents Sea, although the mean chlorophyll concentrations in the Barents Sea observed during the same periods were approximately 2 times those observed in the Greenland Sea (Figures 1b–1d). Given that direct measurements of the abundance and composition of phytoplankton species were not simultaneously undertaken during our study period, we cannot substantiate our conclusion that the Greenland Sea had greater number of DMS-producing phytoplankton species than did the Barents Sea during the study period. Nonetheless, historical field observations confirm that in the Greenland and Barents Seas the major phytoplankton species found during the bloom period were reported to be prymnesiophytes *Phaeocystis pouchetii* and diatoms including *Chaetoceros socialis*, *Skeletonema costatum*, and *Thalassiosira* spp. (Degerlund & Eilertsen, 2010; Sakshaug, 2004). The phytoplankton species differed strikingly in their DMS production capacities. Prymnesiophytes are high DMSP producers and contained DMSP-cleavage enzymes; thereby rapidly transforming DMSP to DMS. By contrast, the diatoms contain cellular DMSP but lack DMSP-cleavage enzymes. Therefore, they produce little DMS (e.g., Keller et al., 1989; Park, Lee, Shin, Jeong, et al., 2014; Stefels et al., 2007). In particular, the prymnesiophytes *Phaeocystis pouchetii* was reported to be a dominant species in terms of DMSP and DMS productions in the Greenland Sea during the spring bloom period (Galí & Simó, 2010), exhibiting an intracellular DMSP concentrations in *Phaeocystis* 10–100 times that of diatoms (Hatton & Wilson, 2007; Stefels et al., 2007).

The positive correlation between the atmospheric DMS mixing ratio and the air mass exposure to chlorophyll is surprising and indicates that DMS emissions from the Arctic Ocean during the bloom periods over the three years of observation were largely governed by DMS-producing phytoplankton (containing cellular DMSP and the DMSP cleavage enzyme). However, in typical marine environments, low-DMS producing phytoplankton (e.g., diatoms and nanoeukaryotes) are generally abundant. In such environments, other complex processes (including grazing activity, bacterial degradation of dissolved DMSP, and cell lysis) are major contributors to DMS production (Calbet & Landry, 2004), and the correlation between atmospheric DMS and air mass exposure to ocean biology may deviate considerably from a simple positive correlation.

To further confirm our conclusion, we estimated the seawater DMSP concentration based on the chlorophyll concentration and the vertical mixing regime (Galí et al., 2015) (Text S1 for more information). As planktonic DMSP is the main precursor of DMS in the surface ocean, the DMSP to chlorophyll ratio is a good measure of the relative contribution of the DMS-producing phytoplankton to total phytoplankton biomass. During our observation periods (May in 2010, 2014, and 2015), the DMSP to chlorophyll ratio in the Greenland Sea

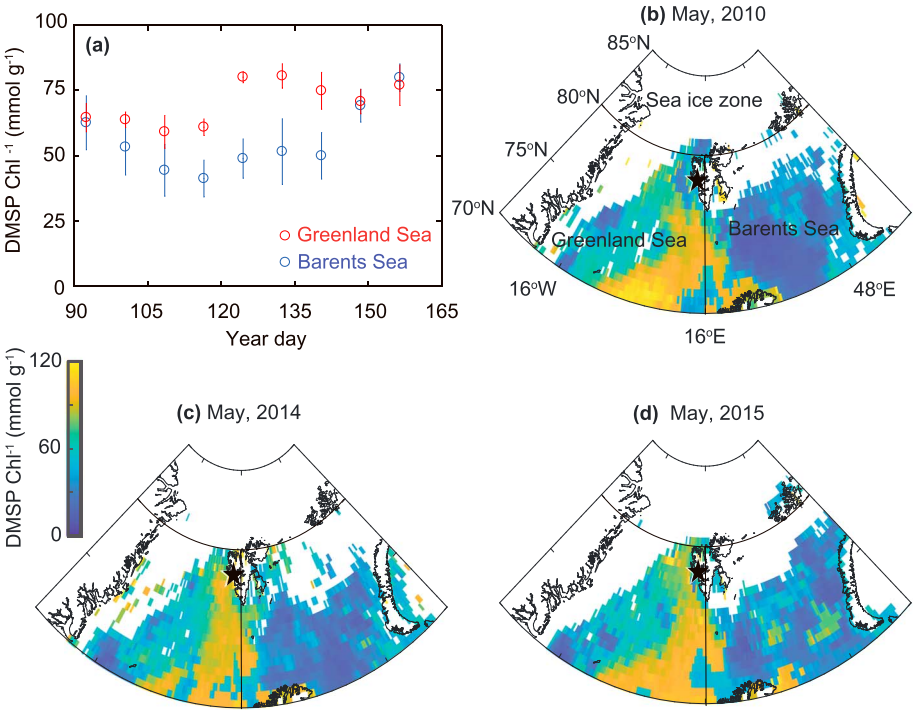

Figure 3. (a) The 8 day mean dimethyl sulfide (DMSP) to chlorophyll ratios for May in 2010, 2014, and 2015. The blue and red circles indicate the mean DMSP to chlorophyll ratio for the Barents Sea (70°N–80°N, 16°E–50°E) and the Greenland Sea (70°N–80°N, 25°W–16°E), respectively. The error bars indicate 1 standard deviation (1σ) from the mean values obtained in 2010, 2014, and 2015. Monthly mean DMSP to chlorophyll ratio during May in (b) 2010, (c) 2014, and (d) 2015. The stars indicate the location of the Zeppelin station (78.5°N, 11.8°E), Svalbard.

was 50% greater than that in the Barents Sea (Figure 3). Application of this method to ocean regions near Svalbard for the period 2006–2015 also revealed that the DMSP concentration was higher in the Greenland Sea compared with the Barents Sea (see Figure 3 in Heintzenberg et al., 2017). Both direct observations and diagnostic model results are consistent with our finding that the DMS production capacity of the Greenland Sea was higher than that of the Barents Sea during phytoplankton bloom periods in the Arctic Ocean.

Laboratory and field studies revealed that variations in the atmospheric DMS mixing ratio are highly correlated with variations in light intensity: higher light intensity induces greater oxidative stress on oceanic phytoplankton, and thereby greater DMS production (Galí et al., 2013; Toole & Siegel, 2004; Vallina & Simó, 2007). Moreover, a higher wind speed at the air-sea interface will drive greater DMS release from a given ocean domain (Zemmelink et al., 2004). As a result, any difference in the intensity of solar radiation and the wind speed at air-sea interface could change DMS production and its release to the atmosphere (see Text S1 for more information). In both the Greenland and Barents seas, solar radiation increased, whereas wind speed decreased during the transition from April to May. However, we did not find any discernable differences in these two parameters in the Greenland and Barents Seas (Figure S7). Consequently, the high DMS production capacity of the Greenland Sea inferred from our analysis was likely to be associated with a greater abundance of prymnesiophytes containing high DMSP content and DMSP cleavage enzyme, whereas the low DMS production capacity of the Barents Sea appeared to be associated with a lower abundance of DMS-producing phytoplankton but a greater abundance of other phytoplankton producing less DMS, including diatoms.

3.4. Possible Enhancement of DMS Production in the Marginal Ice Zone

The marginal ice zone is biologically dynamic and commonly associated with intense phytoplankton blooms (Oziel et al., 2017). Therefore, the dynamic sea ice edge blooms of phytoplankton may have impacts on DMS production and its subsequent release into the Arctic atmosphere (Hayashida et al., 2017). Several field

studies have reported high DMS and DMSP concentrations near the Arctic marginal sea ice zone (e.g., Matrai & Vernet, 1997; Matrai et al., 2007). We evaluated possible enhancement of DMS production near the marginal sea ice zones in the Greenland and Barents seas. In this analysis we determined whether the air mass back trajectories that passed over the sea ice margins (15–80% ice-covered regions, as defined by Stroeve et al., 2016 and Strong & Rigor, 2013) corresponded to elevated atmospheric DMS mixing ratios. The results of this analysis did not indicate that DMS levels were elevated for those air masses sweeping through the boundary layer over the sea ice margins (Figure S8). The absence of enhancement of DMS production at the sea ice margins does not necessarily indicate the absence of such a mechanism, as our analysis method was not sufficiently sensitive to detect signs of elevated DMS production and its subsequent release. More elaborate analyses will be needed to investigate the possibility of enhanced DMS production at the sea ice margins.

4. Conclusions and Implication

A total of 2,141 atmospheric DMS mixing ratios measured above the remote Arctic Ocean during phytoplankton bloom periods over 3 years were analyzed in conjunction with data on the exposure of air masses reaching the observation site (Svalbard) to ocean biology. A comparison of the atmospheric DMS mixing ratios measured at Zeppelin with the air mass exposure to ocean biology of the Arctic Ocean region adjacent to Zeppelin provided insights into the DMS production capacity of the Arctic Ocean. The approach described here is a potentially useful tool for detecting changes in the regional and temporal DMS production capacity of the Arctic Ocean.

In the future, a decrease in sea ice coverage across the Arctic Ocean may increase the annual primary production (Arrigo et al., 2008). Moreover, phytoplankton blooms may occur at progressively earlier times during a season (Kahru et al., 2011). Such increases in the ocean primary productivity and an earlier onset might profoundly change the oceanic production of DMS and its eventual release into the atmosphere (Levasseur, 2013). The DMS production capacity of the ocean depends critically on the phytoplankton species composition and the complex food web mechanisms (Stefels et al., 2007).

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