

Seasonal variations of sulfur aerosols at Zhongshan Station, East Antarctica

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Abstract Observations of atmospheric methane-sulfonic acid (MSA) and non-sea-salt sulfate (nss-SO_4^{2-}) from December 2010 to November 2011 at Zhongshan Station are presented in this paper. MSA and nss-SO_4^{2-} average concentrations were $24.2 \pm 37.9 \text{ ng}\cdot\text{m}^{-3}$ ($0.5\text{--}158.3 \text{ ng}\cdot\text{m}^{-3}$) and $53.0 \pm 82.6 \text{ ng}\cdot\text{m}^{-3}$ (not detected [n.d.] – $395.4 \text{ ng}\cdot\text{m}^{-3}$), respectively. Strong seasonal variations of MSA and nss-SO_4^{2-} , with maxima in austral summer and minima in winter, were examined. The high concentrations of sulfur compounds in December may be attributed the dimethyl sulfide (DMS) emissions from the marginal ice zone, when open water near the sampling site was important in impacting the sulfur species of January and February at Zhongshan Station. In austral winter, there was almost no phytoplanktonic activity in offshore waters, and atmospheric sulfur compounds likely had long-range transport sources.

Keywords MSA, nss-SO_4^{2-} , seasonal variations, East Antarctica, Zhongshan Station

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1 Introduction

The dimethyl sulfide (DMS) oxidation byproducts methane-sulfonic acid (MSA) and nss-SO_4^{2-} in the atmosphere are believed to contribute to cloud condensation nuclei (CCN) number concentration, alter atmospheric albedo, and regulate the climate (the CLAW hypothesis)^[1-2]. In contrast to the Northern Hemisphere, atmospheric sulfur aerosols in Antarctica are natural rather than anthropogenic^[3]. Therefore, it is very important to study marine biogenic sulfur emissions at high southern latitudes, where such emissions are expected to strongly dominate the natural sulfur cycle.

However, because of the harsh environment in Antarctica, most sulfur aerosol studies are performed in austral spring and summer^[4-7]. Seasonal and annual observations are still sparse^[8-10]. It has been demonstrated that there is a strong seasonal cycle of sulfur aerosols in Antarctica. These variations are related to phytoplanktonic activity and

sea ice condition variation in the Southern Ocean^[8]. This productivity could be enhanced following sea ice retreat^[11] because of the release of micronutrients (such as iron)^[12-13] and algae^[14-15] by sea ice melt. In addition, most of the seasonal sea ice zone (south of 60°S) is covered by sea ice with low phytoplanktonic activity and DMS emissions^[16-17]. Significant DMS concentrations and emissions have been detected in the polynyas or open waters off coastal regions of Antarctica^[18-20]. Therefore, attention should be paid to linking phytoplanktonic activity variations in such polynyas with atmospheric sulfur aerosols in coastal regions of Antarctica.

In this study, we present seasonal variations of sulfur aerosols from December 2010 through November 2011. The seasonal variation of sulfur species is discussed. The influence of oceanic phytoplanktonic activity on variations of these species is also analyzed through remote sensing technology. We hope that this research provides information for analysis of the source of the species in aerosol at coastal regions of Antarctica.

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2 Experiment

2.1 Sampling site

Zhongshan Station (69°22'S, 76°22'E) is a Chinese Antarctic research base to the southeast of Prydz Bay in East Antarctica (Figure 1). That bay is the third largest surrounding Antarctica, and the purpose of sulfur aerosol observation at Zhongshan Station was to investigate the influence of marine phytoplanktonic activity in the bay on atmospheric sulfur species. The sampling site was on a hill facing upwind of the station, to avoid local contamination.

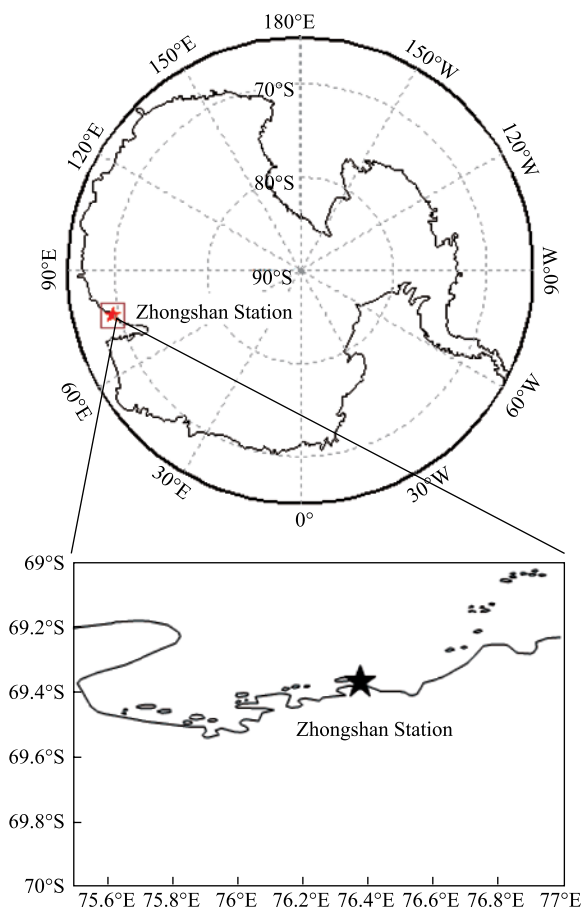


Figure 1 Sampling location.

2.2 Sampling and chemical analyses

Sampling and measurement methods were the same as described in Xu et al.^[21] and Chen et al.^[22]. Aerosol samples were collected with a Whatman 41 filter using a high-volume bulk sampler Model M241 (University of Miami, USA). The sampling interval was 10 d with flow rate $\sim 1 \text{ m}^3 \cdot \text{min}^{-1}$. After sampling, sample filters were stored in a refrigerator at 4°C. A total of 36 high-volume bulk samples were collected from December 2010 through November 2011. A Dionex ICS-2500 ion chromatograph (IC) was used to analyze aerosol samples for water-soluble ions, including MSA, SO_4^{2-} , Cl^- ,

Na^+ , and Mg^{2+} . Experimental methods were as follows. About 1/8 portion of each filter was placed in 50 mL of deionized water, ultrasonicated for 40 min, and leached overnight. Then, the sample solutions were injected into the IC system through 0.22- μm filters. Cations were analyzed with a CS12A analytical column and CG12A guard column with the MSA eluent, and anions were analyzed with an AS18 analytical column and AG18 guard column with KOH eluent. Detection limits were 0.13 $\text{ng} \cdot \text{m}^{-3}$ for MSA, 0.016 $\text{ng} \cdot \text{m}^{-3}$ for SO_4^{2-} , 0.38 $\text{ng} \cdot \text{m}^{-3}$ for Na^+ . Precision of the analytical procedures based on seven spiked samples was $< 5\%$.

2.3 Calculation of nss-SO_4^{2-}

Nss-SO_4^{2-} is commonly calculated by the equation $[\text{nss-SO}_4^{2-}] = [\text{SO}_4^{2-}]_{\text{total}} - [\text{Na}^+] \times 0.252$, where 0.252 is the mass ratio of $\text{SO}_4^{2-}/\text{Na}^+$ in seawater. However, when using this equation, negative nss-SO_4^{2-} values may result, especially during austral fall and winter. The reasons for this were discussed by Hall and Wolff^[23], the most important of which was the fractionation of sulfate aerosols from the formation of $\text{Na}_2\text{SO}_4 \cdot 10 \text{H}_2\text{O}$ when temperature was $< -8.2^\circ\text{C}$ ^[24]. Therefore, in consideration of this fractionation, we used $\text{SO}_4^{2-}/\text{Na}^+$ ratio 0.13 as in Preunkert et al.^[8] to calculate nss-SO_4^{2-} during winter (April through October) in Antarctica.

2.4 Chl *a* concentrations and meteorological data

Eight-day average Chl *a* concentration distributions in the sampling site sector were obtained from <http://oceancolor.gsfc.nasa.gov> (Modisa L3SMI Chl *a* data, with resolution 4 km \times 4 km). Chl *a* concentrations in selected regions were also calculated for analyzing the distribution of phytoplankton activity. Meteorology data were obtained from Zhongshan Station meteorology observatory operated by the Polar Research Institute of China. Three main parameters, air temperature, wind direction and wind speed, were selected and averaged daily for investigation.

3 Result and discussion

3.1 Ion concentrations

Figure 2 presents ion concentrations from December 2010 through November 2011. Mean concentrations of MSA, nss-SO_4^{2-} , SO_4^{2-} and Na^+ were $24.2 \pm 37.9 \text{ ng} \cdot \text{m}^{-3}$ (0.5–158.3 $\text{ng} \cdot \text{m}^{-3}$), $53.0 \pm 82.6 \text{ ng} \cdot \text{m}^{-3}$ (n.d.–395.4 $\text{ng} \cdot \text{m}^{-3}$), $129.6 \pm 112.8 \text{ ng} \cdot \text{m}^{-3}$ (18.5–416.7 $\text{ng} \cdot \text{m}^{-3}$), and $443.4 \pm 409.5 \text{ ng} \cdot \text{m}^{-3}$ (30.6–1648.2 $\text{ng} \cdot \text{m}^{-3}$), respectively.

In contrast with other sites (Table 1), MSA mean concentration during austral summer at Zhongshan Station was consistent with observation at Halley Station (January 2004 through February 2004) and Dumont d'Urville Station (December through February of 1996/1997, 1999/2000 and 2001/2002), but nearly two times smaller than at Palmer Station (January 1994 through February 1994) and Halley

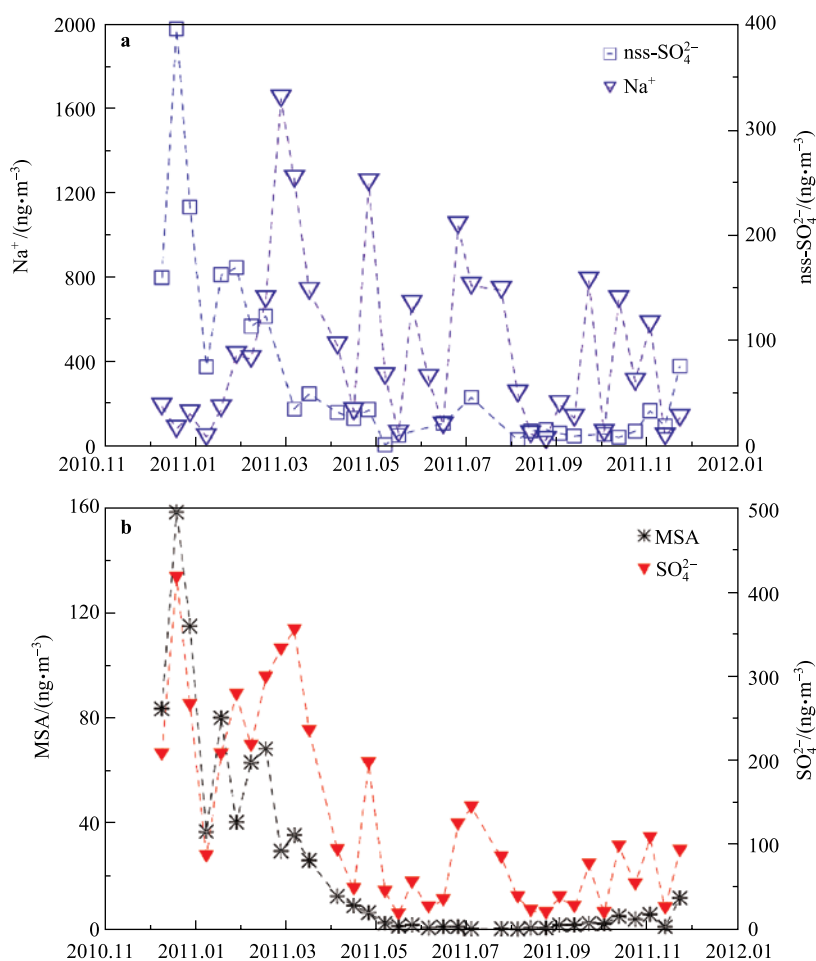


Figure 2 Atmospheric ions concentrations over Zhongshan Station.

Table 1 Summary of average MSA and nss-SO₄²⁻ concentrations observed at various coastal Antarctic sites

Site	MSA/(ng·m ⁻³)	nss-SO ₄ ²⁻ /(ng·m ⁻³)	Month	Reference
Palmer Station 67.77° S, 64.05 °W	180	296	Jan 1994/Feb 1994	[4]
Halley Station 75°35' S, 26 °19'W	83.2	141.3	Jan 2004/Feb 2004 Jan 2005/Feb 2005	[25]
Dumont d'Urville Station 66°40' S, 140°01'E	35	182	Dec–Feb, 1997/1998, 2000/2001, 2002/2003	[8]
	73	247	Dec–Feb, 1996/1997, 1999/2000, 2001/2002	
Zhongshan Station 69°22' S, 76 °22'E	85.5	189.3	Dec–Feb, 2010/2011	This study
	6.6	16.6	Mar–Nov, 2011	

(January 2005 through February 2005). Slightly different than the MSA, nss-SO₄²⁻ mean concentrations in summer at Zhongshan Station were lower than at Palmer (January 1994 through February 1994) and Dumont d'Urville (December through February 1996/1997, 1999/2000 and 2001/2002). Sulfur species variations between the sites could be attributed to biology, sea ice, oxidation and transportation processes, and meteorological conditions.

3.2 Seasonal variations of MSA and nss-SO₄²⁻

The atmospheric sulfur species MSA and nss-SO₄²⁻ exhibited a strong seasonal cycle with summer maxima (Figure 2). The variability of Na⁺ was different from sulfur compounds and had more fluctuation over the year. This may also be attributed to different sources or mechanisms, such as sea spray and biogenic components, formation mechanisms,

various atmospheric reactions, and different size distributions and transport processes^[26].

Figure 2 shows that maximum MSA and nss-SO₄²⁻ values appeared at the end of December, slightly earlier than previous studies at Dumont d'Urville^[5,8-9], in which the maxima sulfur species occurred in January. The maxima of MSA and nss-SO₄²⁻ concentrations were 158.3 ng·m⁻³ and 395.4 ng·m⁻³, respectively. Such high concentrations of sulfur species were possibly related to biogenic emissions of DMS in the coastal regions^[27-28] or marginal sea ice zone^[29-30] of Antarctica, because phytoplankton blooms are frequent there in austral spring and summer. Monthly means of sulfur species are shown in Figure 3, from December through March. Both MSA and nss-SO₄²⁻ decreased rapidly, by a factor of 4–10. Their concentrations decreased from 121.1 ng·m⁻³ to 30.8 ng·m⁻³ and 277.6 ng·m⁻³ to 28.5 ng·m⁻³, respectively. From April through October, very low MSA sulfur species concentrations were observed. During that period, because the ocean was covered by ice and there was very weak solar radiation, primary productivity was very low in the Southern Ocean. This led to weak DMS emissions, with almost no contribution to sulfur aerosols at Zhongshan Station. It has been reported that sulfur species during winter in coastal Antarctica regions are mostly transported from mid or low latitude regions^[9-10] because the lifetimes of MSA and nss-SO₄²⁻ could be as long as a few weeks^[31]. However, MSA and nss-SO₄²⁻ showed a rapid increase from October to November. In contrast to the value at the beginning of November, MSA rapidly increased by a factor of 12. However, the increase of monthly average MSA values relative to winter was not obvious. This may be attributed to missing sample data at the end of that month. The high concentrations of MSA and nss-SO₄²⁻ may be attributed to the early spring phytoplankton bloom in the coastal region or marginal sea ice zone, which followed the sea ice retreat^[17,19] and increase of solar radiation^[32]. In addition, we found a strong relationship between MSA and nss-SO₄²⁻ ($R^2 = 0.91$, $n = 28$, $p < 0.01$), which is the reason that they had a common precursor (DMS).

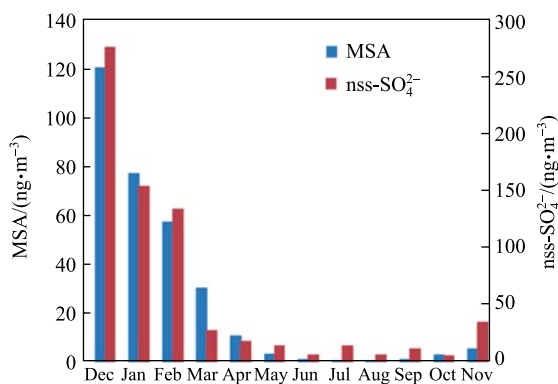


Figure 3 Monthly average MSA and nss-SO₄²⁻ concentrations at Zhongshan Station.

3.3 Influences on MSA and nss-SO₄²⁻ seasonal variation

Because the only source of MSA is believed to derive from the oxidation of DMS^[33] and DMS production is largely related to phytoplanktonic activity, it is reasonable to link the sulfur species to that activity in the oceans. A study by Legrand and Pasteur^[10] demonstrated that marine source regions affecting the sulfur aerosol cycle in high-latitude southern regions were primarily south of 50°S, with an increasing contribution of emissions from regions south of 60°S in summer^[9]. However, most regions in the sea ice zone (south of 60°S) are covered by sea ice with weak biological activity and DMS emissions^[16]. Conversely, as much as hundreds of nmol·L⁻¹ DMS concentrations have been found in polynyas along the coastal regions of Antarctica, such as the Ross and Amundsen seas^[17-18,34]. Thus, it is of interest to investigate locations with major influences on the sulfur species at Zhongshan Station. To discuss the influence of oceanic DMS emissions on atmospheric sulfur compounds, attention should be given to ice-free open waters with high primary productivity. As shown in Figure 4, the dominant daily wind direction was northeast in spring and summer at Zhongshan Station. Because the sampling site was very close to open water (< 1 km), the direct air mass source is offshore.

Figure 5 (bottom panels) depicts relatively strong phytoplanktonic activity in November, i.e., spring in the Southern Hemisphere, following sea ice retreat. The contributions of DMS emissions from the marginal ice zone would be much greater than those from open water offshore of the sampling site, because the ice-free open water area was small with little phytoplanktonic activity (~0.1 mg·m⁻³). However, in general, because both that activity and ice-free water area were not substantial, in combination with relatively low air temperature and solar radiation that could possibly impact the DMS oxidation process^[35], the sulfur aerosol concentration was low.

In December 2010, high Chl *a* concentration was observed along 62°S. In the waters near Zhongshan Station (65°S–70°S, 70°E–85°E), biological activity was low through the end of December (Chl *a* average concentrations were 0.2221–0.8286 mg·m⁻³) (Figure 5). However, the low Chl *a* mean values there did not coincide with the high concentrations of MSA (mean 121.3 mg·m⁻³, range 83.9–158.3 mg·m⁻³) and nss-SO₄²⁻ (mean 277.6 mg·m⁻³, range 159.8–359.4 mg·m⁻³) in December. This suggests that the high concentration of sulfur species in December could hardly have originated from the nearby ocean but were possibly derived from the air mass over the marginal ice zone (along 62°S). It has been reported that in the marginal ice zone of the Weddell Sea, Chl *a* concentration and primary productivity could be as high as 3.0 mg·m⁻³ and 490 mg·C·m⁻²·d⁻¹ in austral spring^[36]. Therefore, as ocean with weak biological DMS emissions offshore of the sampling site, DMS emissions from highly productive oceans to the north (following the sea ice retreat around 62°S) are important in impacting sulfur species

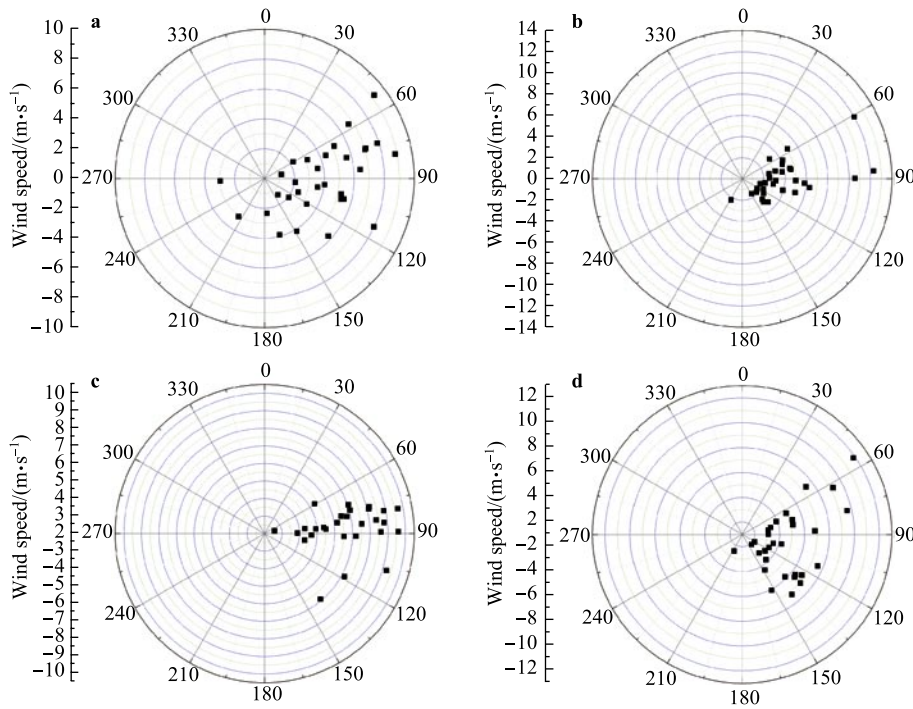


Figure 4 Daily wind rose diagram for spring and summer. **a**, December 2010; **b**, January 2011; **c**, February 2011; **d**, November 2011.

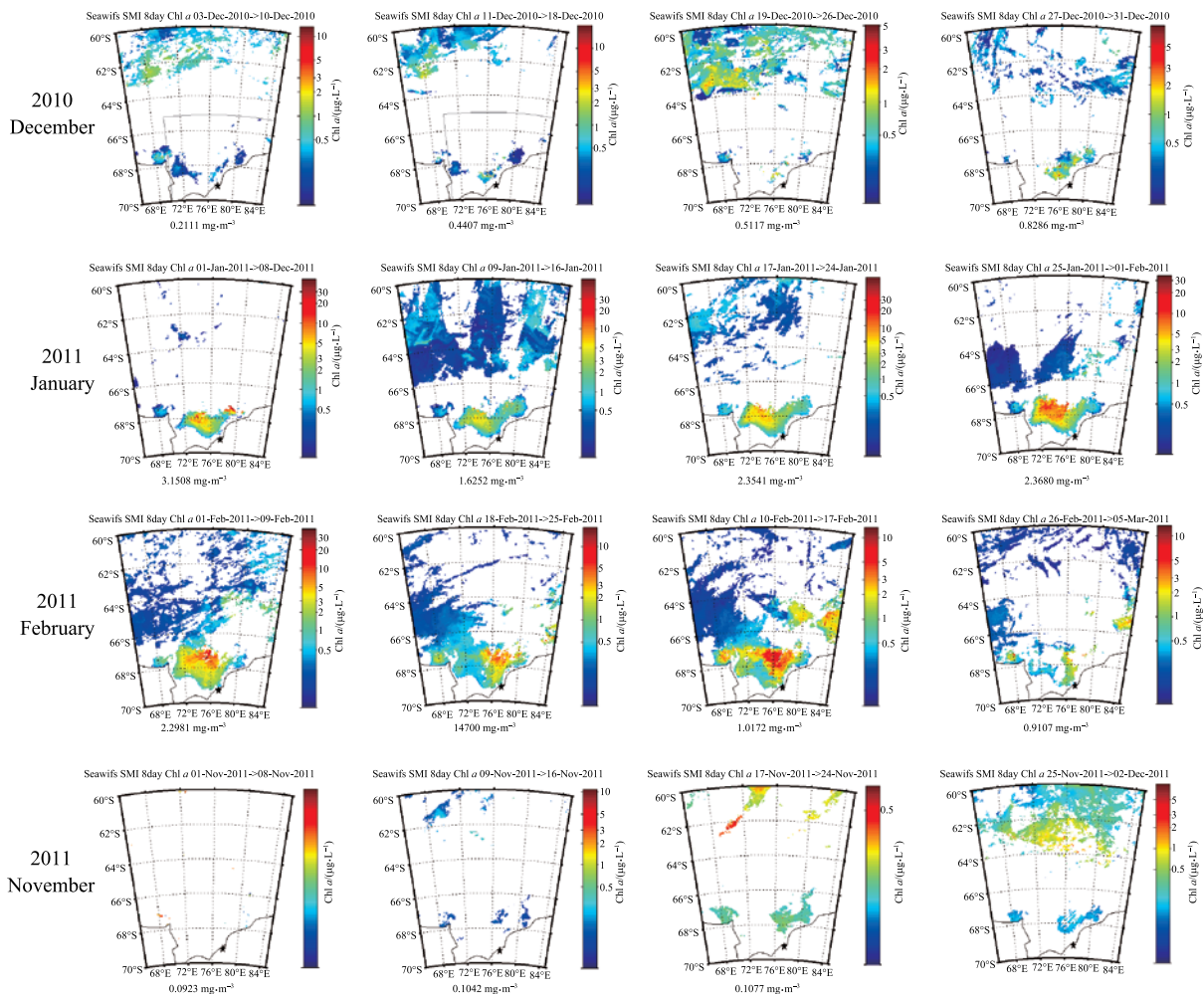


Figure 5 Eight-day average Chl *a* concentrations in various months. Values under each map are Chl *a* mean concentrations in the area off Zhongshan Station (65°S–70°S, 70°E–85°E).

in coastal regions of Antarctica.

From early January to February, Chl *a* concentration along 60°–64°S began to decline (Figure 5). However, a very high biomass level in surface sea water was detected near the sampling site (Chl *a* concentrations were 1.0172 mg·m⁻³ to 3.1508 mg·m⁻³; Figure 5), and phytoplankton bloom always followed sea ice melt. In addition, the average Chl *a* concentration in ice-free open water offshore of the sampling site decreased rapidly from the beginning of January, together with an increase of open water area, through the end of February. Therefore, the dynamics of sea ice would significantly affect phytoplanktonic activity^[37], further modulating DMS emissions. The maximum Chl *a* value in early January coincided with high atmospheric MSA and nss-SO₄²⁻ concentrations (115.2 mg·m⁻³ and 226.8 mg·m⁻³, respectively), suggesting that DMS emission from the adjacent ocean directly influenced the sulfur aerosols. Park et al.^[38] reported that atmospheric DMS mixing ratios at Svalbard (78.5°N, 11.8°E) were strongly correlated with variability in Chl *a* concentration in nearby waters ($r = 0.89$). Because the lifetime of atmospheric DMS in mid austral summer is close to one day^[25], phytoplankton blooms in water offshore of the sampling site could account for the high sulfur species concentrations at Zhongshan Station. However, the high Chl *a* values were not always consistent with the high concentrations of MSA (Figures 2 and 5). For example, a high MSA concentration in late February (63.7 mg·m⁻³, Figure 2) corresponded with a low Chl *a* concentration (1.0172 mg·m⁻³, Figure 5). This could be attributed to the substantial open water area. In late summer, the ice-free open water area maximized, which could also lead to release of abundant DMS from the ocean to atmosphere. In addition, the air mass at Zhongshan Station may be impacted by air mass transport from inland, which could contain low sulfur species concentrations^[7]. The low temperature and rapid horizontal mixing may also affect the formation of sulfur species in February. Therefore, sulfur species concentrations in that month were not high.

Beginning the end of February, sea ice began to freeze, and Chl *a* concentration in waters offshore of the sampling site was at a low level (Figure 5). However, phytoplanktonic activity could also have been enhanced during sea ice formation^[11]. The large value of MSA (36 ng·m⁻³) in early March would be a response to that. During austral winter (April through October), local emission of biogenic DMS would be weak because of heavy sea ice coverage and lack of sunlight, with almost no phytoplanktonic activity. Sulfur species could potentially be transported from mid or low latitude regions. Minikin et al.^[9] stated that DMS byproducts in coastal regions of Antarctica originated from between 50°S and 60°S in austral winter. Therefore, in that season, sulfur species at Zhongshan Station possibly originated from mid or low latitudes, with high primary productivity via long range transport.

4 Summary

The data collected at Zhongshan Station from December

2010 through November 2011 indicated that atmospheric MSA and nss-SO₄²⁻ concentrations had a strong seasonal variation, with maxima in austral summer and minima in winter. Variations of sea ice conditions and phytoplankton activity in offshore waters were important in affecting sulfur species concentrations over Zhongshan Station. However, given a lack of high-resolution observation of those species (sampling intervals were 10 d in the study), we could not analyze in detail the species variations. In addition, to fully understand the impact of DMS emissions from offshore water on those variations at Zhongshan Station, the atmospheric DMS mixing ratio and seasonal variation of seawater DMS concentration must also be investigated. Therefore, improvements in sulfur cycle study at Zhongshan Station are required in the future.

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