

Significant Underestimation of Gaseous Methanesulfonic Acid (MSA) over Southern Ocean

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Supporting Information

ABSTRACT: Methanesulfonic acid (MSA), derived from the oxidation of dimethylsulfide (DMS), has a significant impact on biogenic sulfur cycle and climate. Gaseous MSA (MSA_o) has been often ignored in previous studies due to its quick conversion to particulate MSA (MSA_p) and low concentrations. MSA_o, MSA_p, and nss-SO₄²⁻ were observed simultaneously for the first time with high-timeresolution (1 h) in the Southern Ocean (SO). The mean MSA_g level reached up to 3.3 ± 1.6 pptv, ranging from ~24.5 pptv in the SO, contributing to $31\% \pm 3\%$ to the total MSA (MSA_T). A reduction of the MSA to nss-SO₄²⁻ ratios by about 30% was obtained when MSA_{σ} was not accounted for in the calculation, indicating that MSA, was very important in the assessment of the biogenic sulfur contributions in the



atmosphere. Mass ratios of MSA to nss-SO₄^{2–} increased first and then decreased with the temperature from -10 to 5 °C, with a maximum value at the temperature of -3 °C. Positive correlations between MSA_g to MSA_T ratios and temperature were presented, when the temperature was higher than 5 °C. This study highlights the importance of MSAg for understanding the atmospheric DMS oxidation mechanism and extends the knowledge of MSA formation in the marine atmosphere.

INTRODUCTION

Methanesulfonic acid (MSA) and nonsea-salt sulfate (nss- SO_4^{2-}), derived from the oxidation of dimethylsulfide (DMS), have an important impact on cloud condensation nuclei (CCN) in the marine boundary layer (MBL),^{1,2} resulting in the changes of radiation budget and climate.³⁻⁶ However, some studies argued that there was limited evidence for the link between DMS and CCN in the MBL, according to the observations and modeling studies in the last decades.^{7,8} Hence, an understanding of the products of DMS oxidation and how they interact with marine particles is important to understand how DMS impacts CCN. Different from nss- SO_4^{2-} , MSA is formed exclusively from the oxidation of DMS,⁶ which provides a unique tracer for estimating the amount of biogenic nss-SO4²⁻ from DMS oxidation.¹⁰

MSA and $nss-SO_4^{2-}$ are often associated with submicron particles in the marine atmosphere.^{9,11} Biogenic sulfur products (MSA and $nss-SO_4^{2-}$) have been investigated in Antarctica.¹²⁻¹⁴ Long sampling intervals of even several days were required to accommodate the detection limits,^{15,16} as filtered sampling methods were used in previous studies.^{12,15} However, the oxidation of DMS occurs rapidly in the marine atmosphere (lifetime of approximately 1-2 days).^{17,18} In this case, it is difficult to examine the chemical processes of DMS using long interval observations. Particulate MSA (MSA_p) was

often investigated, 15,16,19,20 as gaseous MSA (MSA_{σ}) was expected to be converted to MSA_p quickly in the atmosphere. Hence, the investigation of products from the DMS oxidation was mainly based on MSA_p in previous studies.^{9,10,20,21} MSA_σ data was rare and often ignored in the atmosphere, as MSA_a was scarcely measured. However, modeling simulation results showed that high levels of MSAg were presented in marine atmosphere.²² The contribution of MSA to other biogenic sulfur compounds was significantly underestimated without MSAg. Hence, MSAg, as well as other biogenic sulfur compounds should be determined to better understand the marine biogenic sulfur cycle and climate effect.

In this study, $MSA_{g'}$ MSA_{p} , and $nss-SO_4^{\ 2-}$ were observed simultaneously for the first time with high-time-resolution (1 h) in the Southern Ocean (SO). Observations were carried out over a large range of latitudes from 43°S to 78°S and longitudes from 63°E to 175°W. The results in this study provided new insight into the formation of MSA_g and MSA_n in the SO, as well as the impact of MSA_g on the marine biogenic sulfur cycle.

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EXPERIMENTS AND METHODS

The R/V "Xuelong" Cruise and Observation Regions. Observations were made on-board the R/V "Xuelong", which, in leg I, sailed from Christchurch, New Zealand, on November 27, 2017 and arrived in the Ross Sea on December 4, 2017, as indicated in Figure 1. The cruise tracks in Figure 1 as well as



Figure 1. Cruise tracks of the observation in the SO.



Figure 2. Spatial distributions of MSA_g and MSA_p . (a) MSA_p levels during leg I ($ng \cdot m^{-3}$). (b) MSA_g levels during leg I (pptv). (c) MSA_p levels during leg II ($ng \cdot m^{-3}$). (d) MSA_g during leg II (pptv).

the spatial distributions of MSA_g and MSA_p in Figure 2 were created with Ocean Data View.^{23,24} The vessel left the Ross Sea on December 15, 2017 and arrived in Prydz Bay on December 28, 2017. Leg II was begun from Prydz Bay on January 4, 2018 and ended back in the Ross Sea on January 16, 2018 (Figure 1). R/V "Xuelong" left for Christchurch on February 11, 2018 and then arrived at Christchurch on February 16, 2018. The detailed observation information is summarized in Table S1.

Monitoring System and Air Sampling. An In-situ Gas and Aerosol Compositions monitoring system (IGAC, Model S-611, Machine Shop, Fortelice International Co. Ltd., Taiwan, http://www.machine-shop.com.tw/) and a Single Particle Aerosol Mass Spectrometer (SPAMS, Hexin Analysis Instrument Co. Ltd., China) were used to determine the gases and aerosols water-soluble ions, and particle chemical compositions, respectively (Figure S1). Note that the major contamination sources were from the chimney, which was located at the stern of the vessel and about 25 m above the sea level. To minimize the impact of the vessel emissions, the air sampling inlet connecting to the monitoring instruments was fixed to a mast at 20 m above the sea surface, located at the bow of the vessel. A total suspended particulate (TSP) sampling inlet was positioned at the top of the mast. Conductive silicon tubing²⁵ with an inner diameter of 1.0 cm was used to make the connection to the instruments.

Gases and Aerosols Water-Soluble Ion Constituents. Gas and aerosol water-soluble ion constituents were determined using a semicontinuous IGAC monitor. Gases and aerosols were separated and streamed into a liquid effluent for online chemical analysis at an hourly temporal resolution. The analytical design and methodology for the determination of gas and aerosol water-soluble ions have been described in detail in previous studies.^{26–29} The IGAC monitoring system consisted of three main units, including a Wet Annular Denuder (WAD), a Scrub and Impact Aerosol Collector (SIAC), and an ion chromatograph with a sampling flow of 16.7 LPM. The WAD consisted of two concentric Pyrex glass cylinders with a length of 50 cm and inner and outer diameters of 1.8 and 2.44 cm, respectively. The inner walls of the annulus were wetted with ultrapure water (18.2 M Ω /cm). The collection of acidic and basic gases relies on the diffusion and absorption of gases into a downward flowing aqueous solution. The SIAC had a length of 23 cm and diameter of 4.75 cm and was positioned at an angle to facilitate the collection of enlarged particles. Ultrapure water was fed continuously into the nozzle at 1.2 mL/min and heated to 140 °C to vaporize the water. Steam was sprayed directly toward the particle-laden air to improve the humidity of the flue gases. Fine particles were enlarged and subsequently accelerated through a conicalshaped impaction nozzle and collected on the impaction plate. The gas and aerosol liquid samples from the WAD and SIAC were drawn separately by a pair of syringe pumps, where one syringe collected the current sample (55 min) and the other injected the previous sample. The samples were then subsequently analyzed for anions and cations by an online ion chromatography (IC) system (Dionex ICS-3000). The injection loop size was 500 μ L for both anions and cations. Six to eight concentrations of standard solutions were selected for calibration, depending on the target concentration, in which the R^2 was above 0.997 (Figure S2). The detection limits for MSA⁻, SO₄^{2–}, Na⁺, and Cl⁻ were 0.09, 0.12, 0.03, and 0.03 μ g/ L (aqueous solution), respectively. The nss-SO₄²⁻ concentrations were calculated from the following equation

$$[nss - SO_4^{2^-}] = [SO_4^{2^-}]_{total} - K[Na^+]$$
(1)

where K equals 0.252, the weight ratio of sulfate and sodium in seawater.³⁰ The concentrations of sodium and total sulfate ranged from 6.7 to 7210.6 and ~1441.2 ng·m⁻³, respectively (Figure S3).

Metrological and Satellite Data of Sea Ice and Chlorophyll-A. Meteorological parameters such as temperature, humidity, wind speed, and direction were measured using an automated meteorological station deployed on the R/ V "Xuelong".

Remote sensing data was used to show the spatiotemporal distribution of chlorophyll-a and sea ice concentration in the study region. Due to the cloud effect and swath limits, we chose the 8 day data sets for the remote sensing of chlorophyll-a from MODIS-aqua (https://modis.gsfc.nasa.gov) with a spatial resolution of 4 km. Considering the effect of sea ice, we used the sea ice concentration data from the daily 3.125-km AMSR2 data set³¹ (available at https://seaice.uni-bremen.de). Chlorophyll-a data in the region from 160°E to 170°W and



Figure 3. Relationship between MSA to nss-SO₄²⁻ ratios and temperature observed in the SO from November 2017 to February 2018. (a) Mass ratios of MSA_p to nss-SO₄²⁻. (b) Mass ratios of MSA_T to nss-SO₄²⁻.

 60° S to 80° S, during the period from November 27, 2017 to February 20, 2018, were first gridded into a 200 × 136 matrix using the Kriging method in SURFER software (version 7.3.0.35) to obtain full coverage of the study region. Each grid of the gridded data sets with a sea ice concentration less than or equal to 15% was regarded as comprising all water.³² The time series of the total regional mean value in the study region was then plotted.

RESULTS AND DISCUSSION

Spatial Distributions of MSA_a and MSA_b in the SO. Along the leg I tracks, MSA_p concentrations ranged from 14.6 to 392.6 ng·m⁻³, with an average of 45.5 \pm 32.0 ng·m⁻³ (Figure 2a and Table S1). These observations consisted of MSA_p levels obtained during the cruise conducted in Antarctica from October 1992 to January 1993,³³ ranging from 2 to 360 ng·m⁻³. High MSA_p levels (with an average concentration higher than 50 ng·m⁻³) were observed in MA1, MA2, MA3, and MA4 regions, with respective averages of 99.6 \pm 22.9, 84.0 \pm 38.3, 61.7 \pm 16.3, and 57.4 \pm 24.6 ng·m⁻³. MSA_o ranged from ~24.5 pptv, with an average of 5.9 \pm 4.7 pptv (Table S1). Modeling simulation results showed that MSA_o concentrations ranged from 1 to 9.6 pptv over the Antarctica MBL²² and around ~13 pptv over Halley Station from January to March 2004.¹⁸ Similar to the spatial distribution of MSA_{ν} , high levels of MSA_{g} (with an average higher than 5 pptv) occurred in MA1 and MA2, with average concentrations of 19.5 \pm 5.2 pptv and 11.7 \pm 5.1 pptv, respectively.

 MSA_p concentrations ranged from 0.3 to 165.4 ng·m⁻³ during leg II (Table S1), consisting of the observation results reported in the SO and Antarctica.^{12,14,16,20} The highest MSA_p level occurred in MA5, with an average of 100.3 ± 18.6ng·m⁻³. High concentrations of nss- SO_4^{2-} were also observed in this region (Figure S4). However, during leg I, the MSA_p concentrations in this region were low, with an average of 38.1 ± 8.3 ng·m⁻³ only. Variations in MSA_p and nss- SO_4^{2-} concentrations in the Ross Sea were associated with phytoplankton activity in this region (Figure S5) as high atmospheric levels of MSA and nss- SO_4^{2-} were often observed during the summer season with high phytoplankton concentrations.

The mean MSA_p concentration during January and February in MA5 was much higher than the MSA_p concentration observed during the early December in the same region (Figure 2a,c). However, the mean MSA_g level (average of 2.4 \pm 0.6 pptv) in MA5 during leg II was lower than the MSA_g level (average of 7.2 \pm 1.5 pptv) observed in this region during leg I. Contrarily, high levels of MSA_g were presented in MG1 and MG2, with averages of 6.7 ± 2.2 pptv and 5.3 ± 0.3 pptv, respectively, while the MSA_p levels were low in MG1 and MG2. These results indicated that the variations in MSA_g levels were not always consistent with MSA_p levels.

Influence of MSA_q on the Results of MSA to $nss-SO_4^{2-}$ **Ratios.** The mass ratio of MSA to $nss-SO_4^{2-}$ provides an important insight into the oxidation routes of DMS^{35,36} and the contribution of marine biogenic and other sources to the global sulfur budget.^{9,37} The nss-SO₄²⁻ is mainly derived from marine biogenic sources in high-latitude oceanic regions. Hence, the ratio of MSA to $nss-SO_4^{2-}$ can be used to assess marine biogenic products at high latitude regions.³⁷ The calculated nss-SO₄²⁻ levels ranged from ~1420.0 ng·m⁻³ during the observation periods (Figure S4). A strong positive correlation between mean ratios of MSA_p to nss-SO₄²⁻ (R_p) and temperature $(r^2=0.76)$ was presented, when the temperature was lower than -3 °C (Figure 3a). But the R_p value decreased slightly with increasing temperature from -3 to 5 $^{\circ}$ C. $R_{\rm p}$ values ranged from 0.10 to 0.33 at the temperature between -3 to 5 °C, which were lower than those reported by Berresheim³⁸ ($R_p = 0.78$ in Mar–Apr, 1987) and Pszenny³ $(R_p = 0.54 \text{ in Mar}-\text{Apr, 1989})$ in Antarctica but consistent with the observations reported by Chen¹⁶ $(R_p = 0.23)$ in the SO. In addition, a similar trend of decreasing R_p with increasing temperature $(T > 5 \, ^{\circ}C)$ was also noted in this study, which was in good agreement with the observations for temperatures ranging from 5 to 25 °C.^{10,16} The mean R_p value decreased from 0.49 to 0.07 as the temperature increased from 5 to 18 °C, consistent with a R_p value of 0.41 reported by Berresheim⁴⁰ (1990) and 0.2 reported by Aers⁴¹ (1991).

The R_p value initially increased with temperature and then decreased in the temperature range from -10 to 5 °C, with the maximum R_p value occurring in the temperature range from -5 °C to -2 °C (Figure 3a), indicating that ratios of MSA to nss-SO₄²⁻ did not always show an inverse relationship with temperature. The favorable formation of MSA from the oxidation of DMS at low temperature has been confirmed in previous studies;¹⁰ however, high R_p values during the warm season were also reported at Cape Grim.³⁹ Note that these results were obtained at temperatures higher than -5 °C.^{10,16} Hence, the results in this study provide a new understanding of the linkage between temperature.

The relationship between mass ratios of total MSA (MSA_T = MSA_g + MSA_p) to nss-SO₄²⁻ (R_T) and temperature is also illustrated in Figure 3b. Variations in the MSA_T to nss-SO₄²⁻ ratios were similar to those noted for R_p in Figure 3a. However, the R_T value was much higher than the R_p value. The mean R_T value ranged from 0.14 to 0.51 for temperatures below -3 °C,

which was about 40% higher than the mean R_p value, while for temperatures between -3 and 5 °C, the mean R_T value ranged from 0.18 to 0.47, which was 44% higher than the R_p value. For temperatures above 5 °C, however, the R_T value (ranging from 0.13 to 0.83) was about 70% higher than the R_p value. These observations were attributed to the re-evaporation of MSAg from particles in the atmosphere at high temperature (Figure 4). A strong positive correlation was presented between R_p and



Figure 4. Formation and sources of MSA_g and MSA_p in the marine atmosphere.

 $R_{\rm T}$ ($r^2 = 0.98$), with a slope of 0.69 (Figure S7). It indicates that MSA_g has a significant impact on the ratio of MSA to nss-SO₄²⁻. In this study, mass ratios of MSA to nss-SO₄²⁻ were underestimated by about 30% without considering the MSA_g.

Formation of MSA_g and MSA_p in the Marine Atmosphere. Potential oxidation reaction routes of DMS with different radicals, such as OH, NO₃, BrO, and ClO, have been described in previous studies.⁴² MSA has a much higher saturation vapor pressure than H₂SO₄. Consequently, MSA_g is expected to have a greater atmospheric lifetime compared with that of H₂SO₄ in gas phase,⁴³ which means a more complicated behavior of MSA_g in the atmosphere. Two alternative routes for the production of MSA have been identified in the marine atmosphere (Figure 4). The formation processes described here for MSA were similar to previous studies;^{18,22} however, the production rates of MSA_g and MSA_p have not been provided in previous studies due to the lack of a MSA_g data set in the SO.^{18,44}

Generally, MSA_g and MSA_p are also impacted by transport sources, except for local sources in the atmosphere (Figure 4). MSA_p is attributed to the multiple reactions of DMS (k_1 and k_3), the conversion of MSA_g (k_5), and transport input. MSA_p levels in the atmosphere can be described as follows

$$[MSA]_{g} = k_{1}[DMS]\Delta t_{1} + k_{5}[MSA]_{g}\Delta t_{2} + [MSA_{trans}]_{p}$$
(2)

In comparison with that of MSA_p , the lifetime of MSA_g in the atmosphere is much shorter.⁴¹ MSA_g levels in the atmosphere can be described by the following equation

$$[\text{MSA}]_{\text{g}} = k_2 [\text{DMS}] \Delta t_3 + k_5 [\text{MSA}]_{\text{p}} \Delta t_2 + [\text{MSA}_{\text{trans}}]_{\text{g}}$$
(3)

On the basis of the routes of MSA_g and MSA_p formation described in eqs 2 and 3, respectively, only part of MSA_p is formed directly from MSA_g . This is the reason why MSA_g levels do not always account for the observed levels of MSA_p (Figure S4a and Figure 2).

The lifetime of MSA_g was only about 20–40 min,⁴³ according to the modeling calculation. The Δt_2 for the conversion of MSA_p was significantly shorter than Δt_3 , according to the oxidation of DMS.⁴⁵ The sink of MSA_g was determined by the gas to particle conversion in the atmosphere. Due to very short lifetime, MSA_g was mainly derived from the local oxidation of DMS and scarcely affected by the long-term transport sources. In this case, when there was no transport input for MSA_p, the MSA_g and MSA_p were derived from the local oxidation of DMS. In this case, the ratios of MSAg to MSAp represented the production rates of MSA_{g} and MSA_{p} from the oxidation of DMS in the marine atmosphere. But the ratios of MSAg to MSAp did not account for the actual production rates of MSAg and MSAp from the oxidation of DMS, when MSA_p was affected by the transport input. The transport input of MSA_p would probably decrease the ratios of MSAg to MSAp. Hence, the production rates of MSA_{g} and MSA_{p} would be higher than the observed MSA_{g} to MSA_p ratios.

The observed MSA_g to MSA_p ratios ranged from ~1.17 throughout the campaign (Figure S6), with an average of 0.47 \pm 0.22 (n = 1874). However, as seen in Figure 5c, ratios of



Figure 5. Influence of temperature on MSA_g and MSA_p . (a) Correlation between MSA_g and MSA_p . (b) Impact of temperature on the correlation between MSA_g and MSA_p . (c) Effect of temperature on MSA_g to MSA_T ratios.

 MSA_g to MSA_T mainly fell within the range of 0.1–0.5. Note that ratios of MSA_g to MSA_T showed an obvious increase with temperature (Figure 5c), when the temperature was higher than 5 °C. As the effective volatility of MSA is uncertain, it is still too short to know the condensational behavior of MSA_g . Hodshire⁴⁶ (2019) simulated the condensational behavior of MSA using the GEOS-Chem model, assuming that MSA will condense as it is ideally nonvolatile and semivolatile. Condensed MSA was not allowed to evaporate to the gas phase in the modeling calculation. In this case, the contribution of MSA_g toward condensation on particles was overestimated, assuming idealized semivolatile condensation with no re-evaporation due to conditional changes. The ratios

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of MSA_g to MSA_T would not be changed with the environmental conditions (such as temperature and RH), when the MSA cannot re-evaporate from the particulate phase. However, positive correlations between MSA_g to MSA_T ratios and temperatures were observed in this study (Figure 5), indicating that the condensation and re-evaporation processes of MSA in the atmosphere were reversible. Although the factors controlling the reversible conversion processes between MSA_p and MSA_g were still uncertain, the re-evaporation of MSA_g from the particulate phase was demonstrated.

Influence of Temperature on MSA_g Levels in the SO. Generally, it is very difficult to find a relationship between MSA_g and MSA_p under the full data set. However, when we investigated the relationships in the distinct regions, we found temperature-based regional correlations between MSA_g and MSA_p. A strong positive correlation was observed between MSA_g and MSA_p in the region MA1 (slope = 0.99, r^2 = 0.94, n= 65) with high temperature ($T > 5 \,^{\circ}$ C, Figure 5a,b). Strong relationships between MSA_g and MSA_p also occurred in MA3 and MA4 (slope = 6.68, r^2 = 0.88, n = 55) for temperatures ranging from -3 to 1 $^{\circ}$ C (Figure 5a,b). Another positive correlation between MSA_g and MSA_p was presented in MA5 (slope = 6.36, r^2 = 0.37, n = 112) for temperatures below -5 $^{\circ}$ C (Figure 5b). In other regions, however, we did not find obvious correlations between MSA_g and MSA_p and MSA_p (Figure 5a,b).

In this study, ratios of MSAg to MSAT were used to assess the impact of temperature on the relationship between MSA_g and MSA_p. Initially, MSA_g to MSA_T ratios increased with temperatures from -13 °C to -5 °C and then decreased to minimum levels at 5 °C (Figure 5c). MSAg to MSAT ratios were mainly concentrated on a range of 0.1 to 0.5, with an average of 0.31 \pm 0.03 (n = 1656) for temperatures between -13 and 5 °C. However, the mean MSA_g to MSA_T ratio increased to 0.43 \pm 0.05 (n = 274) for temperatures higher than 5 °C. MSA_g to MSA_T ratios revealed an increasing trend with temperature, indicating that more MSA_g was formed at high temperatures. The condensation of MSAg onto aerosol surfaces was favorable at low temperatures, but MSA, was reevaporated from MSA_p at high temperature. The observation results confirmed the assumption of MSA_g and MSA_p conversation processes mentioned in the above section.

3.5. Implication of MSA_a on the Biogenic Sulfur Cycle and Climate. The results of this study indicated that MSA_o was very important to the biogenic sulfur compounds in the atmosphere, accounting for more than 30% of the MSA_T. Hence, MSA_g could not be ignored when biogenic sulfur chemical processes were investigated. Previous studies found that SO_4^{2-} was more effective in new particle formation (NPF), while MSA was likely to condense onto existing particles.⁴⁷ However, recent studies have shown that MSA may also enhance cluster formation, as each MSA molecule contained within the configuration made the cluster more stable to improve the NPF.⁴⁸ The re-evaporation of MSA from particle phase MSA was observed in the SO. In this case, MSA_g would become very important when it is transported to the MBL where it can enhance NPF. Though NPF occurred infrequently in the MBL over remote oceans,49 relatively frequent nucleation events have been observed in the central Arctic Ocean during summer.⁵⁰ Nevertheless, previous studies have suggested a link between nucleation events and the formation of sulfuric acid in the oxidation of DMS.^{3,4} However, Pirjola⁵¹ (2000) pointed out that homogeneous binary H₂SO₄-H₂O nucleation would not occur under typical

conditions in the MBL, and ternary $H_2SO_4-H_2O-NH_3$ nucleation would only be sufficiently effective to produce observable particles for DMS concentrations in the range of 400 pptv or higher and very low aerosol condensation sinks.

Interestingly, the study by Hoppel⁴⁷ has opened up the possibility for MSA molecules to aid NPF in the MBL. While the oxidation of DMS is tentatively important for nucleation involving MSA molecules, the presence of MSA_g is certainly more important for the growth of submicrometer marine particles to CCN-sizes.⁷ However, the details of this pathway linking biological activity to particle by number and subsequently the number population of CCN are still controversial and should not be seen as a direct connection between DMS and CCN, as our current understanding of the processes controlling or maintaining the CCN population over the remote oceans is limited.

ASSOCIATED CONTENT

S Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.9b05362.

Gaseous and particulate MSA levels in different regions; gases and aerosols monitoring system using in this study; calibration curves of MSA, chloride, sulfate, and sodium for IGAC monitoring system; time series of particulate sodium and sulfate during the observation cruise; time series of MSA_g , MSA_p , $nss-SO_4^{2-}$, and the meteorological parameters during November 2017 to February 2018; spatial distribution of sea ices and chlorophyll-a concentrations; time series of the ratios of MSA_g to MSA_p during the whole cruise; correlation between MSA_g to $nss-SO_4^{2-}$ ratios and MSA_T to $nss-SO_4^{2-}$ ratios (PDF)

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Notes

The authors declare no competing financial interest.

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