Supporting Information

Significant underestimate of gaseous Methanesulfonic Acid (MSA) over Southern Ocean

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The supplementary information contains 10 pages, including 1 table and 7 figures. The table and figure captions are listed as follow:

Tab. S1 Gaseous and particulate MSA levels in different regions

Region	Longitude	Latitude	$MSA_{g(min)}$	$MSA_{g(max)}$	$MSA_{g(Avg.)}$	$MSA_{p(min)}$	$MSA_{p(max)}$	$MSA_{p(Avg.)}$
	(°E)	(°S)	(pptv)	(pptv)	(pptv)	($ng \cdot m^{-3}$)	($ng \cdot m^{-3}$)	($ng \cdot m^{-3}$)
Leg I	76 - 177	43 - 75	-	24.5	5.9±4.7	14.6	392.6	45.5±32.0
Leg II	72 - 185	43 -78	-	13.0	2.4±0.7	0.3	165.4	33.7±24.8
MA1	173	43 - 51	12.7	24.4	19.5±5.2	73.2	167.0	99.6±22.9
MA2	172	64 - 69	3.9	24.5	11.7±5.1	49.5	392.6	84.0±38.3
MA3	125 - 142	63	3.5	5.0	4.2±0.5	50.8	95.0	61.7±16.3
MA4	85 - 93	61.5	2.6	3.3	2.9±0.2	43.3	144.9	57.4±24.6
MA5	170 - 185	68.2 - 77.8	1.4	4.0	2.4±0.6	57.4	165.4	100.3±18.6
MG1	163 - 177	72 - 75	5.0	21.4	6.7±2.2	20.8	75.0	38.4±21.9
MG2	101 - 106	62	4.8	5.7	5.3±0.3	36.7	71.8	45.5±9.6

Fig. S1 Gases and aerosols monitoring system. An underway biogenic gases and aerosols monitoring system were employed on the R/V "Xuelong" to carried out the observation in the SO. An In-situ Gas and Aerosol Composition monitoring system was used to determine the gaseous and aerosol water-soluble ions. A Single Particle Aerosol Mass Spectrometer was used to determine the particle size distribution and chemical compositions.

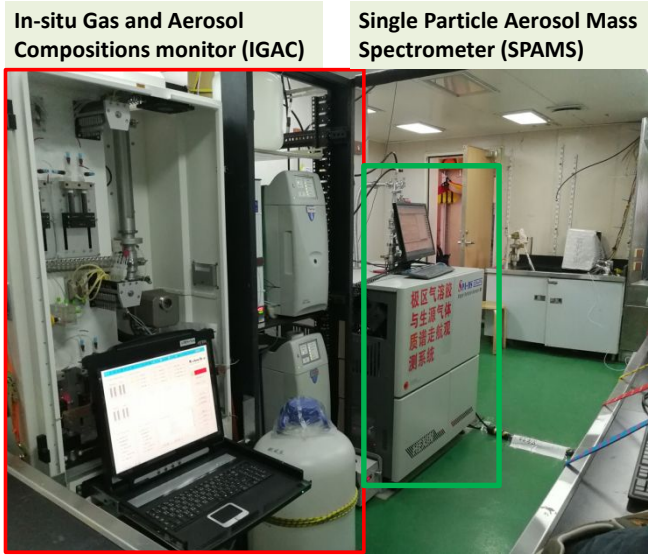


Fig.S2 Calibration curves of MSA, chloride, sulfate and sodium for IGAC monitoring system. (a) Six out of eight concentrations of standard solutions (0.1-1000 ug/L) were selected for MSA calibration (r^2=0.998); (b) Six out of eight concentrations of standard solutions (0.1-2000 ug/L) were selected for Chloride calibration (r^2=0.997); (c) Six out of eight concentrations of standard solutions (0.1-4000 ug/L) were selected for Sulfate calibration (r^2=0.997); (d) Six out of eight concentrations of standard solutions (0.1-2000 ug/L) were selected for Sodium calibration (r^2=0.998).

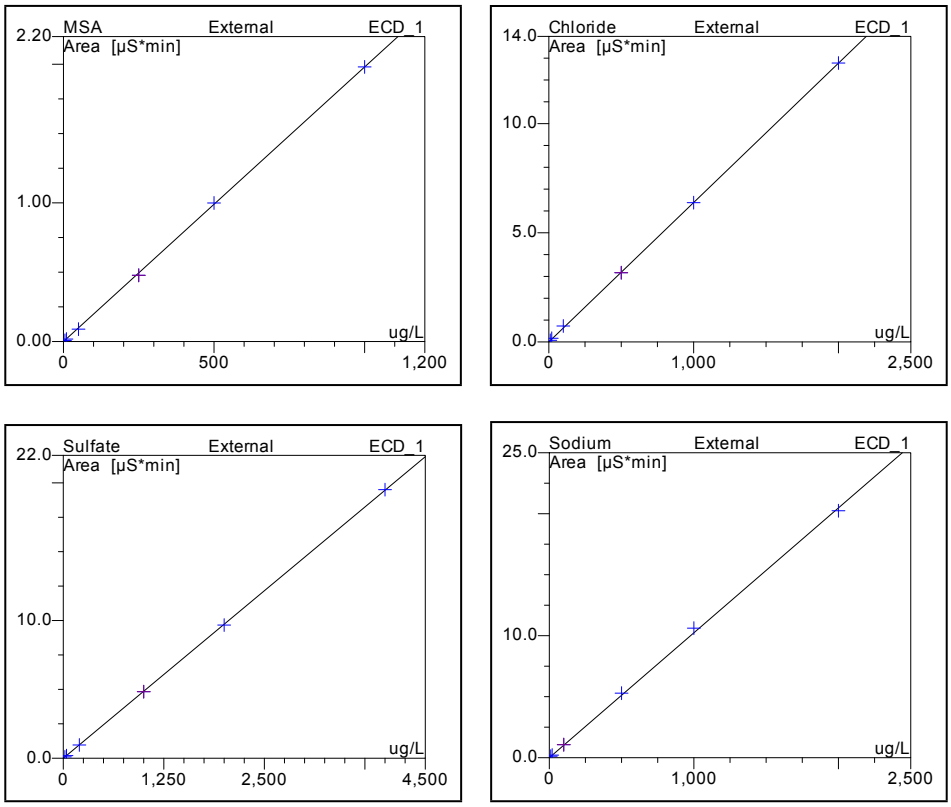


Fig. S3 Time series of particulate sodium and sulfate during the observation cruise.

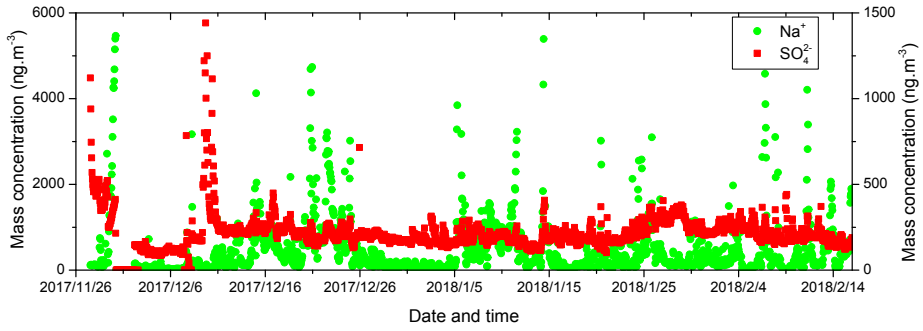


Fig. S4 Time series of gaseous and particulate MSA, nss-SO_4^{2-}, and meteorological parameters during Nov. 2017 to Feb. 2018. (a) Temporal distributions of gaseous and particulate MSA; (b) Temporal distribution of nss-SO_4^{2-}; (c) Temperature and RH; (d) Wind speed and directions.

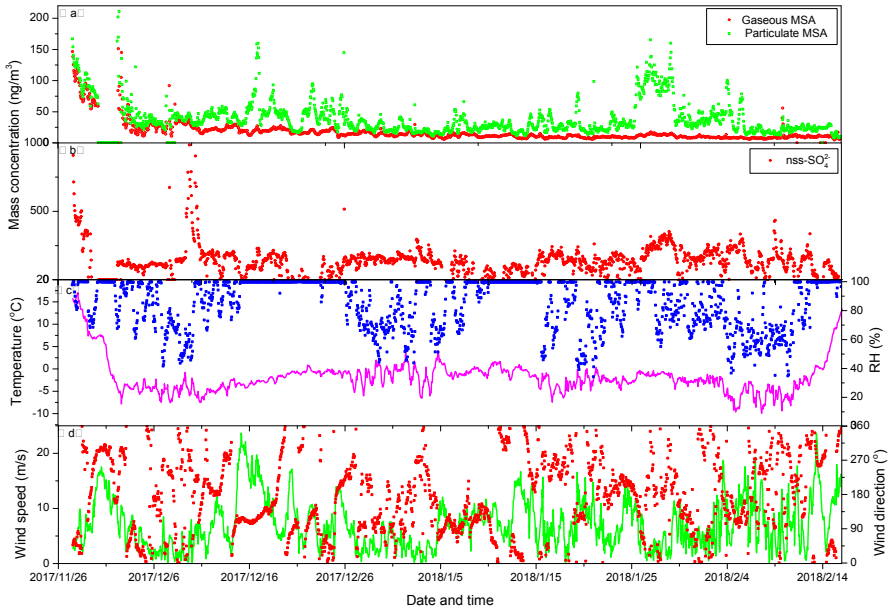


Fig. S5 Spatial distributions of sea ice and Chl-a concentrations. (a) Average sea ice during 4 to 14 December, 2017; (b) Average sea ice during 25 January to 4 February, 2018; (c) Mean Chl-a concentrations during 4 to 14 December, 2017; (d) Mean Chl-a concentrations during 25 January to 4 February, 2018. The Spatial distributions of sea ice and Chl-a concentrations in this figure were created with Ocean Data View (Ref.S1 and S2).

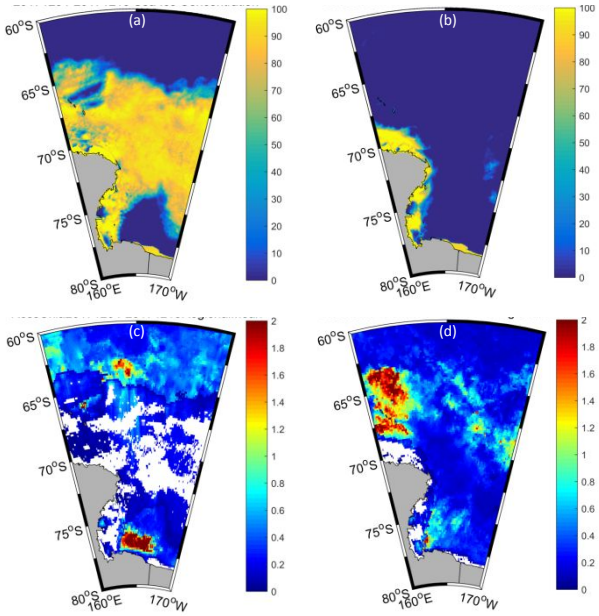


Fig. S6 Time series of the ratios of MSA_g to MSA_p during the whole cruise.

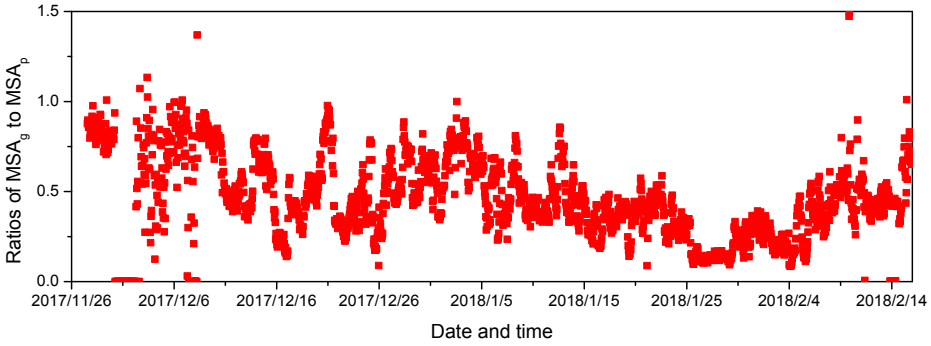
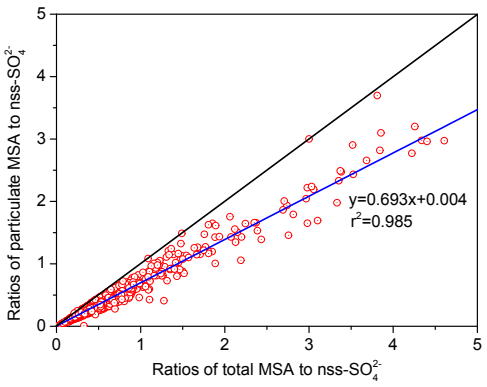


Fig. S7 Correlation between MSA_g to nss-SO_4^{2-} ratios and total MSA_T to nss-SO_4^{2-} ratios. An intensity positive correlation between R_p and R_T (r^2= 0.985) was observed, with a slope of 0.693. The ratios of MSA to nss-SO_4^{2-} were reduced by about 30 % without MSA_g.



References

S1. Schlitzer, R. Ocean Data View, odv.awi.de, 2015.

S2. Schlizer, R. Interactive analysis and visualization of geosciences data with Ocean Data view. *Computers Geosciences*. 2002, 28, 1211-1218.