

## Longitudinal distribution of biogenic sulfur compounds in atmospheric aerosols over the subarctic North Pacific

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Atmospheric aerosols affect the Earth's radiation balance through absorbing and scattering solar radiation directly or acting as cloud condensation nuclei. It is well known that sulfate contribute most to the negative radiative forcing by aerosols. Therefore, elucidating the behavior of sulfur compounds in the atmosphere is important to understand the global climate change. Biogenic sulfur compounds such as non-seasalt (nss-) sulfate and methanesulfonic acid (MSA) are formed by the oxidation of dimethylsulfide (DMS) which produced by marine phytoplankton. These compounds are especially important in the open ocean where the influence of anthropogenic substances is small. In this study, the longitudinal distribution of sulfur compounds in the aerosols over the subarctic North Pacific during summer was shown, and environmental factors determining the distribution were discussed.

Aerosol samples were collected during R/V MIRAI MR07-04 cruise from 24 July to 3 September 2007. The cruise sailed the subarctic North Pacific from Sekinehama, Japan to Dutch Harbor in Alaska, Unites States. Atmospheric aerosols were classified into coarse (diameter  $>2.5 \mu\text{m}$ ) and fine (diameter  $\leq 2.5 \mu\text{m}$ ) modes and collected on Teflon filters by a dichotomous virtual impactor. The major ionic species and MSA in the aerosol samples were measured by ion chromatography.

The mean concentrations of nss-sulfate and MSA during the cruise were  $0.29 \pm 0.18$  and  $0.03 \pm 0.02 \mu\text{g m}^{-3}$ , respectively. Relatively high concentration of ammonium ion was observed only off the coast of Hokkaido, therefore, most of the observed ocean area was considered to be less impacted by anthropogenic pollutants. The concentration of MSA in the east side of the international date line (IDL) were twice of those in the west side. The spacial variation of MSA concentration was similar to that of DMS in both seawater and atmosphere observed in the same cruise. The high concentrations of DMS in the atmosphere could be caused by high wind speed in the east side of IDL. Furthermore, composition of suspended particles including phytoplankton debris showed remarkable increase in coccolith in the east side of IDL, indicating that the change in dominant species of phytoplankton also influenced MSA formation.

Although about 95% of MSA was existed in fine mode, the coarse mode fraction of MSA were higher in the east side than the west side of IDL. This suggests that seasalt, which were mainly existed in coarse mode, increased due to the high wind speed, and a part of MSA adsorbed to the seasalt. On the other hand, the size fraction of nss-sulfate was relatively constant regardless of the ocean area. The concentration of nss-sulfate was lower in the east side of IDL. One possible interpretation is that the increase of seasalt particles caused adsorption of  $\text{SO}_2$  to the particles and removal to sea surface. Since the residence time of seasalt in coarse mode is much shorter than fine-mode aerosols, the contribution of DMS-derived biogenic sulfur compounds to cloud condensation nucleation could be overestimated in the environment with abundant seasalt particles. This study highlights the role of seasalt as a scavenger on the spatial distribution of biogenic sulfur compounds in the marine aerosols.

Keywords: Marine aerosol, Sulfate, Methanesulfonic acid, Dimethyl sulfide, Seasalt