Autocatalytic conversion of oceanic dimethyl sulfide emissions into cloud condensations nuclei affecting the Earth’s albedo

The oxidation of biogenic dimethyl sulfide (DMS) emissions is a global source of cloud condensation nuclei. The amounts of the nucleating $\text{H}_2\text{SO}_4$ (g) species produced in such process, however, remain uncertain. Hydrophobic DMS is mostly oxidized in the gas-phase into $\text{H}_2\text{SO}_4$ (g)+DMSO (g) (dimethyl sulfoxide), whereas water-soluble DMSO is oxidized into $\text{H}_2\text{SO}_4$ (g) in the gas-phase but into $\text{SO}_4^{2-}$+MeSO$_3^-$ (methane sulfonate) on water surfaces. Thus, $R=\text{MeSO}_3^-/\text{non-sea-salt-SO}_4^{2-}$ ratios would therefore gauge both the strength of DMS sources and the extent of DMSO heterogeneous oxidation if $R_{\text{het}}=\text{MeSO}_3^-/\text{SO}_4^{2-}$ for DMSO(aq)+•OH(g) were known. Here, we report that $R_{\text{het}}=2.7$, a value obtained from online electrospray mass spectra of DMSO (aq)+•OH (g) reaction products, which quantifies the MeSO$_3^-$ produced in DMSO heterogeneous oxidation on aqueous aerosols for the first time. On this basis, the inverse $R$-dependence on particle radius in size-segregated aerosol collected over Syowa station and Southern oceans is shown to be consistent with the competition between DMSO gas-phase oxidation and its mass accommodation followed by oxidation on aqueous droplets. Geographical $R$ variations are thus associated with variable contributions of the heterogeneous pathway to DMSO atmospheric oxidation, which increase with the specific surface area of local aerosols.

Biography

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