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World Conference on

Climate Change

October 24-26, 2016 Valencia, Spain

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'Sizing' heterogeneous chemistry in the conversion of gaseous dimethyl sulfide to atmospheric particles

The oxidation of biogenic dimethyl sulfide (DMS) emissions is a global source of cloud condensation nuclei. The amounts of the nucleating $H_2SO_4(g)$ species produced in such process, however, remain uncertain. Hydrophobic DMS is mostly oxidized in the gas-phase into $H_2SO_4(g)$ +DMSO(g) (dimethyl sulfoxide), whereas water-soluble DMSO is oxidized into $H_2SO_4(g)$ in the gas-phase but into SO_4^{-2} + MeSO₃- (methane sulfonate) on water surfaces. Thus, R=MeSO₃-/non-sea-salt-SO42- ratios would therefore gauge both the strength of DMS sources and the extent of DMSO heterogeneous oxidation if Rhet = MeSO₃-/SO₄²- for DMSO(aq) + \cdot OH(g) were known. Here we report that Rhet=2.7, a value obtained from online electrospray mass spectra of DMSO (aq) + \cdot OH(g) reaction products, which quantifies the MeSO₃- 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

Agustín J Colussi is a Senior Research Scientist at CALTECH since 1998 and has published more than 200 papers in environmental physical chemistry.

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