雲核生成の鍵となるジメチルスルホキシドとOHラジカルの不均一反応の研究

Experimental study on the heterogeneous reaction of gaseous OH radical with aqueous DMSO: Determination of the $CH_3SO_3^-$/ SO_4^{2-} production ratio

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The oxidation of dimethyl sulfide (DMS) emitted from ocean (~ 45 Tg S per year) is a global source of cloud condensation nuclei. 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 SO_4^{2-} + $CH_3SO_3^-$ (methane sulfonate) on water surfaces. R = $CH_3SO_3^-/SO_4^{2-}$ ratios therefore indicate the extent of DMSO heterogeneous oxidation if R_{het} = $CH_3SO_3^-/SO_4^{2-}$ for DMSO(aq) + ·OH(g) were determined. Here, products and intermediates of the oxidation of aqueous DMSO initiated by gas-phase hydroxyl radicals, OH(g), at the air-water interface were directly detected by mass spectrometry in a novel setup under various experimental conditions. Exposure of millimolar DMSO aqueous microjets to ~ 10 ns OH(g) pulses from the 266 nm laser flash photolysis of O_3(g)/O_2(g)/H_2O(g)/N_2(g) mixtures yielded an array of interfacial intermediates/products, including $CH_3SO_3^-$ and HSO_4^-, that were unambiguously and simultaneously identified in situ by mass spectrometry. We determined R_{het} = 2.7 from the heterogeneous OH-oxidation of DMSO on aqueous aerosols for the first time. The nearly quantitative production of H_2SO_4(g) (that leads to SO_4^{2-}) in the oxidation of DMSO in the gas-phase versus the R_{het} ~ 2.7 value determined at the air-water interface means that R = $CH_3SO_3^-$/nss-SO_4^{2-} variations in the aerosol, particularly in remote locations, should arise from the competition between the gas-phase versus the heterogeneous DMSO oxidation pathways. The present study reveals that interfacial OH-oxidation processes play a more significant role in the generation and growth of atmospheric aerosol over ocean than previously envisioned.

キーワード：エアロゾル、光化学的エイジング、気候変動、不均一反応、海洋化学
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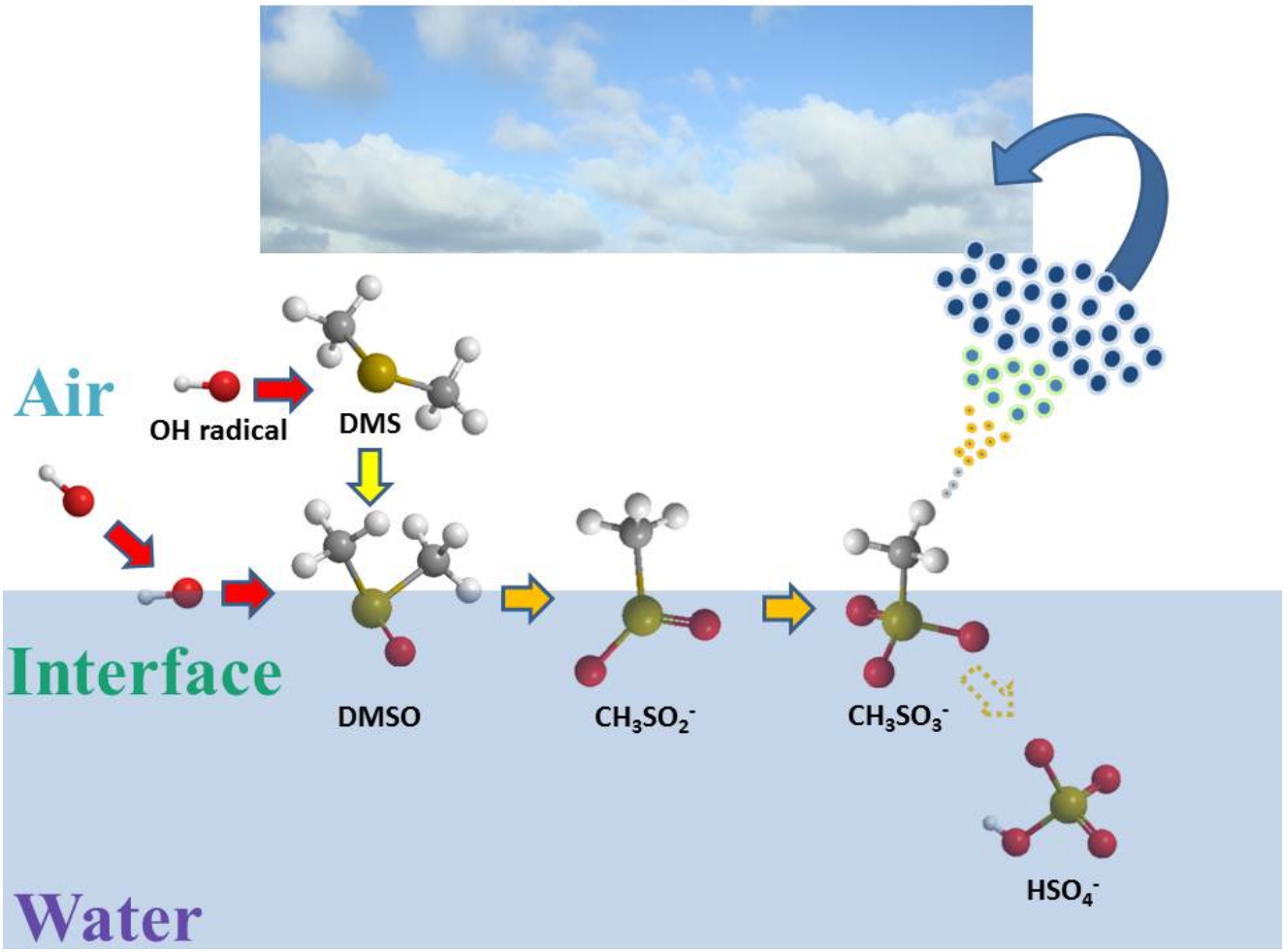

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