Organosulfate Formation in Proxies for Aged Sea Spray Aerosol: Reactive Uptake of Isoprene Epoxydiols to Acidic Sodium Sulfate

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Abstract
Oxidation of isoprene, the biogenic volatile organic compound with the highest emissions globally, is a large source of secondary organic aerosol (SOA) in the atmosphere. Organosulfates, particularly methyltetrol sulfates, formed from acid-driven reactions of isoprene epoxydiols (IEPOX), a key oxidation product, are important contributors to SOA mass. To date, most studies have focused on organosulfate formation on ammonium sulfate particles at low pH. However, recent work has shown sea spray aerosol (SSA) in the accumulation mode (~100 nm) is often quite acidic (pH ~ 2) and IEPOX-derived organosulfates have been identified in marine environments. Herein, we demonstrate that substantial SOA, including organosulfates, are formed on acidic sodium sulfate particles (pH = 1.3), representative of marine aerosol heterogeneously reacting with H2SO4 to form Na2SO4. For acidic sodium and ammonium sulfate particles, 31 and 28% (±1%), respectively, of inorganic sulfate is incorporated into organosulfate species, even though acidic particles with sodium versus ammonium as the primary cation formed 5% (±0.2) less SOA volume and 45% (±6%) less methyltetrol sulfates, suggesting other organosulfates may form. Even though both exhibited core-shell morphology after IEPOX uptake, physicochemical differences were observed via Raman microspectroscopy, with organosulfates identified in both the core and shell of acidic ammonium sulfate SOA particles, but only in the core for acidic sodium sulfate SOA via Raman microspectroscopy. Our results suggest that isoprene-derived SOA formed on aged SSA is potentially an important, but underappreciated, source of SOA and organosulfates in marine and coastal regions and could modify SOA budgets in these environments.

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