2.041 The Effects of Aerosol-Phase State and Chemical Composition on Multiphase Chemistry Leading to Isoprene-Derived Secondary Organic Aerosol Formation.

Early Career Scientist

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Abstract:

Multiphase reactions between gas- and particle-phase constituents are an important formation mechanism for atmospheric secondary organic aerosol (SOA). Aerosol phase state is thought to influence the reactive uptake and multiphase chemistry processes of gas-phase precursors by altering their diffusion rates within particles. This laboratory study systematically examines the reactive uptake probability of isoprene-derived epoxydiols (IEPOX) onto acidic ammonium sulfate particles with selected pre-existing SOA coatings by coupling a flow tube reactor with an iodide-adduct high-resolution time-offlight chemical ionization mass spectrometer (HR-ToF-CIMS). A uniform layer of organics is coated onto the inorganic particles using a potential-aerosol-mass (PAM) oxidation flow reactor, confirmed via atomic force microscopy (AFM) and scanning electron microscopy (SEM). Results show that certain pre-existing SOA coatings could significantly reduce the reactive uptake probability of IEPOX, in some cases by nearly an order of magnitude when the coating thickness is only 10 nm.

Particle composition is also analyzed by online and offline analytical techniques, including an aerosol chemical speciation monitor (ACSM), ultra-performance liquid chromatography interfaced to electrospray ionization (ESI) HR-ToFMS, and ESI coupled to ion mobility spectrometry HR-ToF-MS. Results show that the oxidation state and composition of aerosol particles jointly change their phase state and reactive uptake coefficients. A box model combining experimental data with ambient measurements from the 2013 SOAS campaign is used to assess the effects of pre-existing organic coating on IEPOXderived SOA formation. IEPOX-derived SOA within a diurnal cycle is estimated to be reduced by 16-27% due to pre-existing organic coatings.

Our study provides a potential explanation for the discrepancy between model predictions and field measurements of IEPOX-derived SOA from previous studies. The approach used in this study could be more widely applied to other multiphase chemical systems in regional and global scale models to better predict the impact of SOA their climate, human health, and visibility.