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The role of secondary chemistry in atmospheric oxidation systems: anthropogenic perturbations and gas-particle interactions

Beth Friedman
Colorado State University

Abstract:

Secondary chemistry of primary gases and particles is important to the total secondary organic aerosol (SOA) budget in urban and rural regions, with subsequent impacts on human health, air quality, and climate. The impacts of secondary chemistry on two different systems will be presented. First, we present the impacts of anthropogenic perturbations in the form of sulfur dioxide on the oxidation systems of α- and β-pinene to simulate regions of high BVOC emissions co-located with emissions of anthropogenic pollutants. An oxidation flow reactor simulated atmospheric aging by the hydroxyl radical, and high-resolution time-of-flight mass spectrometry was utilized to identify gas-phase oxidation products and changes to the ensemble system as a function of the SO_2 perturbation. Results show that the SO_2 perturbation impacted the oxidation systems of α- and β-pinene, and that these perturbations affected the oxidation systems of α- and β-pinene differently. Secondly, we utilize the same oxidation flow reactor system to study the primary and secondary gas- and particle-phase emissions of an off-road diesel engine. We present the bulk gas- and particle phase oxidation properties as a function of fuel type (biodiesel vs. diesel), fuel load, and photochemical age due to oxidation by the hydroxyl radical. We show that simultaneous fragmentation and functionalization of the gas-phase species occurs with increased hydroxyl radical exposure; the increased particle-phase mass as a function of oxidation reflects the contribution of the lower-volatility gas-phase species to the particle-phase mass.

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For more information please contact Bonnie Slagel, bonnie@ucar.edu, phone 303-497-8318.