4.034 Tracking the evolution of all carbon in the multigenerational oxidation of biogenic organic compounds.

Early Career Scientist

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Abstract:

Atmospheric oxidation processes quickly transform emitted organic compounds into a multiphase, chemically dynamic system of organic aerosol and gas-phase products. A complete understanding of the fate and transformations of organic carbon in the atmosphere therefore requires a time-resolved quantitative description of both gas- and particle-phase carbon, but attempts to understand the evolution of carbon through atmospheric oxidation has in nearly all cases resulted in a large fraction of “missing” or unidentified carbon. We present here results from laboratory oxidation experiments in which a large suite of state-of-the-art mass spectrometric and spectroscopic instrumentation was employed to measure organic compounds across nearly all volatilities and chemical functionalities typically observed in the atmosphere. Photochemical oxidation of common biogenic emissions was studied, spanning hours to days of simulated atmospheric aging, with a focus here on α-pinene. New chemical ionization mass spectrometric instruments allowed for the quantification of low-volatility gases formed through these processes, and characterization of their role in aerosol

growth. Through systematic variation of reaction conditions (e.g. with and without seed aerosol) coupled with measurements of low-volatility compounds, we explore the importance of measurement artifacts, especially vapor deposition to surfaces, on typical measurements and laboratory experiments. By observing all carbon through multiple generations of oxidation, we examine the transitions between particles and lower-volatility gases with unprecedented detail and place them in the context of current simplified (i.e. two-dimensional) model frameworks, providing a more complete understanding of the evolution of organics in the atmosphere as well as an updated assessment of the capabilities and limitations of current atmospheric instrumentation.