

## **Secondary Organic Aerosol Formation From the Heterogeneous Chemistry of Isoprene-Derived Epoxides: Implications for Air Quality, Climate and Public Health**

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Atmospheric fine particulate matter (PM<sub>2.5</sub>, with aerodynamic diameter < 2.5 μm) impacts Earth's climate directly through scattering/absorbing solar radiation and indirectly through aerosol-cloud interactions, and is associated with adverse effects on human health. Globally, the largest mass fraction of PM<sub>2.5</sub> is organic, and is mostly dominated by secondary organic aerosol (SOA) formed from atmospheric oxidation of hydrocarbons. Isoprene (2-methyl-1,3-butadiene, C<sub>5</sub>H<sub>8</sub>) is the most abundant non-methane hydrocarbon emitted into Earth's atmosphere and is derived from terrestrial vegetation. Although recognized as the single largest source of global SOA, the exact manner in which isoprene-derived SOA is formed remains unclear. Improving our fundamental understanding of isoprene-derived SOA is key to improving existing air quality models, especially in the southeastern U.S. where models currently underestimate observations.

By combining organic synthesis, computational calculations, mass spectrometry, smog chamber studies, flow tube studies and field measurements, we show that reactive epoxides, which include methacrylic acid epoxide (MAE) and isomeric isoprene epoxydiols (IEPOX), produced from the photochemical oxidation of isoprene are key to SOA formation. Furthermore, anthropogenic pollutants, such as acidic sulfate aerosol, enhance isoprene-derived epoxides as an SOA source. In the laboratory, we find that the reactive uptake of synthetic IEPOX and MAE standards onto acidified sulfate aerosol yields known isoprene-derived SOA tracers that we measure in PM<sub>2.5</sub> collected from multiple sites across the southeastern U.S. using a suite of online and offline mass spectrometry methods. Notably, IEPOX- and MAE-derived SOA tracers account for a significant mass fraction (10-25%) of fine organic aerosol in this region. Moreover, real-time continuous chemical measurements of fine aerosol made using an Aerodyne Aerosol Chemical Speciation Monitor (ACSM) during summer 2011 and 2013 resolved an IEPOX-oxygenated organic aerosol (IEPOX-OOA) factor when applying positive matrix factorization (PMF) to the organic mass spectral time series. This factor (or source profile) is found to account for upwards of 33% of the fine OA mass and is strongly correlated with known IEPOX-derived SOA tracers and sulfate, and to some extent with aerosol acidity. We have also investigated the uptake kinetics of MAE and *trans*-β-IEPOX using a flow reactor coupled to the high-resolution chemical ionization mass spectrometry and particle sizing techniques. The reaction probability, also called the reactive uptake coefficient, is calculated for MAE and IEPOX on 1-2 component aerosol particles under various aerosol compositions and environmental conditions in order to probe the chemical drivers of the epoxide uptake. New findings on how this aerosol chemistry might impact climate and public health will also be discussed.