SOA derived from isoprene epoxydiols: Insights into formation, aging and distribution over the continental US from the DC3 and SEAC4RS campaigns

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While isoprene-derived SOA formation has been studied extensively in the laboratory, it is still unclear to what extent isoprene contributes to the overall SOA burden over the southeastern US, an area with both strong isoprene emissions as well as large discrepancies between modeled and observed aerosol optical depth. Under low-NO conditions (<100 pptv), the key gas-phase intermediate is believed to be isoprene epoxide (IEPOX), which can be incorporated into the aerosol phase by sulfate ester formation (IEPOX sulfate), direct hydrolysis, or other mechanisms. As first suggested by Robinson et al. (2011), SOA formed by this mechanism (IEPOX-SOA) has a characteristic fragmentation pattern when analyzed by an Aerodyne Aerosol Mass Spectrometer (AMS) with an enhanced relative abundance of the $C_5H_6O^+$ ion (f_{C6H5O}). Using Positive Matrix Factorization (PMF), we have extracted and identified IEPOX-SOA factors for the all the AMS datasets recorded on flights on the NASA DC8 during the DC3 and SEAC[4]RS campaigns. These campaigns both sampled the SE US over the spring of 2012 and the summer of 2013, respectively, allowing insight into the seasonal pattern of IEPOX-SOA formation. The PMF factors show good, albeit somewhat variable, correlations with f_{C6H5O} within or downwind of high isoprene emitting areas. Results suggest different uptake products to aerosols vs. clouds. Based on this analysis, on average 26-32% of SOA in the SE US boundary layer during the summer of 2013 was IEPOX-SOA, while spring time values were considerably lower (13%). IEPOX-SOA was also found with significantly lower contributions in isoprene-rich areas in the Western US. These results are placed in the context of multiple ground studies compiled by our group (Hu et al., 2015), which also show considerable variability on IEPOX-SOA composition.

Overall, the highest concentrations of IEPOX-SOA were typically found around 1-2 km AGL, several hours downwind of isoprene sources with high gas-phase IEPOX present. IEPOX-SOA was also detected up to altitudes of 5-6 km, with a clear trend towards more aged aerosol at altitude, likely a combination of chemical aging and physical airmass mixing. The unique instrument package aboard the NASA-DC8 allows us to examine the influence of multiple factors (aerosol acidity, aerosol water content, sulfate mass fraction, isoprene and terpene source strength, and intermediate precursors) to the relative and absolute contribution of IEPOX-SOA to the total OA burden, as well as to its composition including the fraction of IEPOX organosulfate and the different AMS tracer ions.

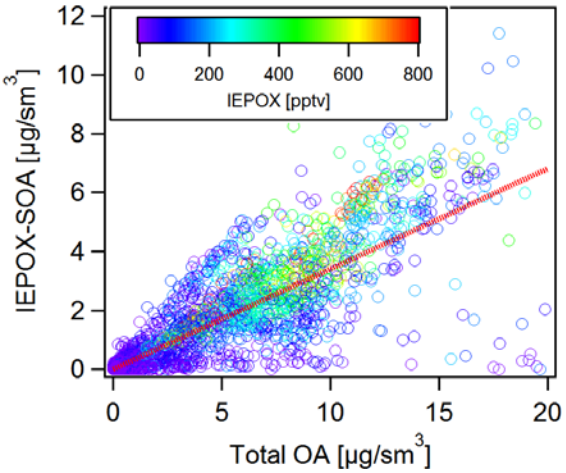

Figure 1. Overview of the contribution of IEPOX-SOA as determined by PMF to the total OA burden in SE US during the SEAC4RS campaign (Summer 2013, 32% Slope).

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