ABSTRACT

Secondary organic aerosol (SOA) constitutes a substantial fraction of fine particulate matter (PM) and has important impacts on climate and human health. The extent to which human activities alter SOA formation from biogenic emissions in the atmosphere is highly uncertain. Southeastern US is an ideal location for studying anthropogenic-biogenic interactions due to high natural emissions and the proximity to anthropogenic pollution sources. We conducted multiple ambient measurements in the greater Atlanta area and Centreville in rural Alabama with a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) during different seasons, as part of the Southeast Oxidant and Aerosol Study (SOAS), post-SOAS study, and Southeastern Center for Air Pollution and Epidemiology (SCAPE) study. We also developed a novel system for direct and online characterization of water-solubility of aerosols by coupling a Particle Into Liquid Sampler (PILS) to the HR-ToF-AMS. Based on integrated laboratory and field measurement results, we provide direct observational evidence on the magnitude of anthropogenic influence ($SO_2$, NOx) on biogenic SOA formation. We show that isoprene-derived SOA is directly mediated by the abundance of sulfate, where 1 mg/m$^3$ decrease in sulfate can result in 0.23-0.42 mg/m$^3$ decrease in isoprene-derived SOA. Anthropogenic NOx is shown to enhance nighttime SOA formation via nitrate radical oxidation of monoterpenes, resulting in the formation of condensable organic nitrates. We find that the majority of OA is water-soluble in both rural and urban sites. The water-solubility of OA factors, resolved with Positive Matrix Factorization analysis of AMS data, is directly investigated for the first time and is found to exhibit different degrees of solubility. Taken together, these comprehensive chemical characterizations of aerosols in the southeastern US demonstrate that anthropogenic sulfate and NOx can mediate 43-70% of total OA (29-49% of total PM$_1$) in summer. The water-solubility of OA factors provides insights for interpretation of different OA subtypes and improves understanding of the complex OA sources in the atmosphere.

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