PhD Dissertation Defense

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Real-Time Chemical Characterization of Atmospheric Organic Aerosol in the Southeastern United States By Aerosol Mass Spectrometry

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The chemical characteristics and sources of atmospheric organic aerosol (OA) in the southeastern United States (U.S.) are least known compared to the other areas due to the complex mixture of anthropogenic and biogenic emissions in this area. The chemical characterization of atmospheric OA requires real-time continuous measurements over different seasons to capture the variability of precursor emissions as well as changes in meteorological conditions. Previous studies in this region were limited by low-time resolution and low-mass identification. Continuous approaches, such as the newly developed Aerodyne Aerosol Chemical Speciation Monitor (ACSM) applied here, have the benefit of high-time resolution and immediate chemical analysis of ambient non-refractory fine aerosol using electron ionization followed by quadrupole aerosol mass spectrometry. The ACSM is designed for long-term monitoring, making it suitable for identifying sources of atmospheric OA based on its chemical composition and temporal variations. This dissertation demonstrates that the ACSM is capable of stable and reproducible operation over extended measurement periods. ACSM measurements compared well with established air-monitoring data in urban Atlanta, GA, and rural Look Rock, TN. Source apportionment of summer OA measured by the ACSM using positive matrix factorization in Atlanta yielded four sources (factors), namely hydrocarbon-like OA (HOA), semi-volatile oxygenated OA (SV-OOA), low-volatility oxygenated OA (LV-OOA), and isoprene epoxydiol (IEPOX)-OA. The IEPOX-OA was found to be strongly correlated (r^2 ~0.6) with known secondary organic aerosol (SOA) tracers and was directly attributed to the heterogeneous chemistry of IEPOX. Measurements at Look Rock also demonstrated significant contributions of IEPOX-OA to the total OA mass (on average 33%). Strong association between the IEPOX-OA and sulfate is consistent between sites. Despite no clear association of IEPOX-OA with locally estimated aerosol acidity and liquid water content, box model calculations of IEPOX uptake accounting for the role of acidity and aerosol water result in predictions of IEPOX-derived SOA tracers at the Look Rock site moderately correlated with observations (r^2 = 0.3 – 0.4).

The remarkable consistency of IEPOX-OA contribution across this region reveals the importance of heterogeneous chemistry of IEPOX in forming OA. Findings from this dissertation could help in developing policy and regulation for mitigating air pollution in this region.

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