Investigating sources and sinks of organic aerosol: NO$_3$-initiated oxidation of isoprene and heterogeneous oxidation of organic aerosol

Thesis by

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Acknowledgments

While only my name appears on the front of this work, it is the result of a collective effort studying a complex system, one involving instrumental development, laboratory and field studies, data analysis, and – as in many human experiences – tedium and drama, frustration and elation. I am honored to have navigated the scientific challenges and emotional vicissitudes with the support of a great team of collaborators and friends.

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Abstract

Secondary organic aerosol (SOA) are important components in atmospheric processes and significantly impact human health. The complexity of SOA composition and formation processes has hampered efforts to fully characterize their impacts, and to predict how those impacts will be affected by changes in climate and human activity. Here, we explore SOA formation in the laboratory by coupling an environmental chamber with a suite of analytical tools, including a gas-phase mass spectrometry technique that is well suited for tracking the hydrocarbon oxidation processes that drive SOA formation. Focusing on the oxidation of isoprene by the nitrate radical, NO₃, we find that reactions of peroxy radicals (RO₂) to form ROOR dimers is an important process in SOA formation. The other gas-phase products of these RO₂ reactions differ from what is expected from studies of simpler radicals, indicating that more studies are necessary to fully constrain RO₂ chemistry. Finally, we examine the role of heterogeneous oxidation as a sink of organic aerosol and a source of oxygenated volatile organic compounds in the free troposphere.
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