



Ali Akherati

Ph.D. Dissertation Defense

12 p.m., Tuesday, April 13, 2021
via Zoom

Advisor: Shantanu H. Jathar

Co-advisor: Jeffrey R. Pierce

Committee: Tami C. Bond, John Volckens, Delphine K. Farmer

Organic Aerosol Modeling of Wildfires

Abstract

Atmospheric aerosols impact climate, air quality, human health, and visibility. Organic aerosols (OA) account for a large fraction of the submicron atmospheric aerosol mass globally, and biomass burning (BB) is the largest contributor to atmospheric OA. BBOA is poorly understood due to uncertainties in the physical and chemical processes that drive its emissions, production, and evolution in the atmosphere. My work combines extensive data analysis of laboratory and field-campaign data with new state-of-the-art OA box modeling to explore the physicochemical evolution of primary OA (POA) and SOA from BB emissions. I use several novel modeling tools to investigate the importance of different SOA precursors in BB laboratory experiments, and I explore the key processes and mechanisms driving the discrepancies between laboratory and field BB measurements.

In the first project, I modeled the SOA production from various precursor vapors in BB emissions during the NOAA FIREX (Fire Influence on Regional and Global Environments Experiment) laboratory campaign. I showed that oxygenated aromatics played an important role in forming a large fraction of the BBSOA measured in laboratory experiments. Accounting for (1) vapor-wall-loss artifacts in Teflon chamber experiments and (2) losses of Intermediate Volatile Organic Compounds (IVOC) species in the transfer tubing during injection of smoke to the chamber resulted in doubling the SOA production in our modeling work. In addition, using fire-temperature-dependent emission profiles and campaign-average emission profiles in modeling sets produced relatively similar results as using emission profiles from each individual fire, showing that fire temperature might be sufficient for characterizing SOA formation in BB plumes.

In the second project, I analyzed measurements of atmospheric BB plumes to explore discrepancies between laboratory and field evolution of OA mass and composition. For this, we developed an OA model that simulated dilution, oxidation chemistry, thermodynamic properties, and aerosol microphysics and applied it to data from the Western Wildfire Experiment for Cloud Chemistry, Aerosol Absorption, and Nitrogen (WE-CAN) field campaign. Our data analyses suggested that SOA precursor emissions, on average, were lower in the WE-CAN field campaign than during the FIREX laboratory measurements. Our model results also showed that dilution-driven evaporation of POA with simultaneous photochemical production of SOA was likely to explain the measured trends in OA mass and composition in the field data. We predicted a rapid transformation of SOA precursors within the first hour after emission that resulted in OA enhancement because of high OH concentrations during this time; however, the airborne WE-CAN measurements miss much of this rapid aging due to limits on how close the plane can fly to the fires. Semi-volatile organics, heterocyclic species, and oxygenated aromatics were the most important SOA precursors in these large wildfire plumes.

My work makes significant contributions to understanding the physicochemical evolution of smoke particles and bridging differences between laboratory and field measurements.