

Ph.D. Defense Announcement

Jakob Lindaas

July 2, 2020 at 10:00 a.m.

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Ph.D. Defense

Thursday, July 2, 2020
10:00 a.m.

Defense
Virtually (see Teams link at bottom)

Post Defense Meeting
Virtually

Committee:
Emily Fischer (Adviser)
A.R. Ravishankara
Jeffrey Collett
Shantanu Jathar (Mechanical Engineering)

Investigating emissions and evolution of reactive nitrogen in western U.S. wildfire smoke plumes

Wildfires are an important source of reactive nitrogen (Nr) to the atmosphere. Nr in smoke contributes to the production of ozone (O₃), the formation of secondary inorganic and organic aerosol, and nitrogen deposition to downwind ecosystems, with attendant negative impacts on human and ecosystem health and important consequences for the earth's radiative budget. Scientific aircraft are able to sample smoke where and when it occurs, making them ideal platforms from which to gather observations of gases and particles in smoke. This defense will present results from three analyses investigating the emissions and evolution of reactive nitrogen in wildfire smoke using in situ aircraft data from the Western wildfires Experiment on Cloud chemistry, Aerosol absorption, and Nitrogen (WE-CAN).

First, I will present a general overview of Nr emissions from 16 wildfires using a suite of chemical and physical measurements made during WE-CAN. I will explore the factors controlling the distribution of Nr in smoke between reduced and oxidized forms, and compare emission factors (EF) with previous literature. I find that reduced N compounds generally make up more than half of the total measured Nr (39 - 80%, median = 66%) and find that both combustion conditions and fuel N content influence the ratio of the sum of measured ammonia and the sum of oxidized nitrogen. Ammonia (NH₃) EFs are similar to or higher than previous lab and field observations, and nitrogen oxide (NO_x) EFs are generally lower than previous estimates.

Next I will explore the evolution of NH₃ in fresh and aged smoke. Focusing on 8 pseudo-Lagrangian sampled plumes, I observe e-folding loss timescales for NH₃ with respect to gas-particle partitioning on the order of minutes to hours, similar to previous estimates in fresh smoke. I find empirical evidence for the association of NH₃ and nitric acid (HNO₃) to form ammonium nitrate (NH₄NO₃), though not all plumes contain conditions favorable to NH₄NO₃ formation. Fresh, dense plumes injected at higher altitudes (and lower temperatures) are more likely to favor NH₄NO₃ formation, a conclusion consistent with previous model simulations in the literature.

Lastly, I will investigate the production of peroxyacetic nitric anhydride (PAN) and peroxypropionic nitric anhydride (PPN) in the same set of pseudo-Lagrangian sampled plumes. PAN and PPN are produced in all smoke plumes sampled, with the production rate similar to the handful of previous observations. I then use a simple observation-based model to determine the dominant precursors of PAN and PPN in fresh smoke plumes. I infer that acetaldehyde is the dominant immediate PAN precursor in large western wildfire smoke plumes, with biacetyl also serving as an important precursor. To my knowledge this is the first time these conclusions have been drawn for smoke from an observational framework. With respect to PPN, I unexpectedly find that at least one other immediate PPN precursor other than propanal is likely needed to explain PPN production, and suggest that this precursor may be ethylglyoxal. However, very few in situ measurements of ethylglyoxal exist to test this hypothesis.

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