Towards a Mechanistic Understanding of Land-Atmosphere Interactions of Reactive Oxides of Nitrogen (DE-SC0014443)

Jonathan D. Raff

School of Public and Environmental Affairs and Department of Chemistry, Indiana University
Bloomington, IN 47405-2204

Global soils are known to be a major source of oxides of nitrogen to the atmosphere. While these emissions have traditionally be been associated with nitric oxide (NO) and nitrous oxide (N_2O), recent laboratory measurements and satellite-global model comparisons suggest that nitrous acid (HONO) and nitrogen dioxide (NO_2) may also be an important constituent of the N-budget. Unfortunately, the processes controlling these emissions are not understood due to challenges in elucidating details of the relevant abiotic and biogenic processes occurring within the terrestrial environment. This is especially true for soil microbial emissions of reactive nitrogen (e.g., NO, NO_2, and HONO)—gases that directly and indirectly affect climate by controlling the oxidative capacity of the atmosphere, lifetime of greenhouse gases, and formation rate of aerosols. In this presentation, I will discuss recent progress made in understanding the abiotic and biogenic processes that determine the fate of HONO in soil. We carried out kinetics studies using a coated wall flow reactor and surface composition studies using nano-DESI and nanoSIMS [at the Environmental Molecular Sciences Laboratory (EMSL)] to investigate the role that minerals and organic matter play in storing and releasing HONO in soil. In addition, mesocosm experiments were conducted to characterize the biogenic mechanisms of reactive nitrogen release from agricultural and urban soil. Flux chamber experiments on agricultural and urban soil samples were carried out to identify soil that emitted reactive nitrogen. The biological nature of these emissions was probed in a series of amendment (NH_4^+ and/or nitrapyrin) studies; 16S rRNA genes and expressed rRNA from the samples were sequenced to provide information on the microbial community composition and activity. Finally, I will describe a newly developed soil flux chamber array that will be used to measure reactive nitrogen fluxes in a northern hardwood forest during a one-month field campaign at the University of Michigan Biological Station in July 2016. This will provide us with the opportunity to field test hypotheses formulated in the laboratory; the results will be used in parameterizations of soil reactive nitrogen flux in future chemical transport models.