Reactions at the Aerosol Surface: Connections between Aerosol Diversity and Reactive Chlorine Chemistry

Kerri Pratt

University of Michigan, USA

Author list (excluding presenting author)

Qianjie Chen, Daun Jeong, Stephen McNamara, Siyuan Wang

Abstract

Heterogeneous reactions play a major role in atmospheric chemistry, impacting oxidant levels and aerosol chemical composition. These reactions are typically simulated in models using parameterizations based upon chemically-resolved aerosol mass concentrations, which assumes homogeneous distribution of chemical components across all particles with complete surface availability for reaction. However, single-particle measurements have shown that the aerosol population is a diverse mixture, suggesting that this is a poor assumption. As an example, the heterogeneous production of nitryl chloride ($ClNO_2$) leads to ozone and particulate matter production, with significant air quality impacts. Laboratory and computational chemistry studies have shown that N_2O_5 reacts at the atmospheric particle surface, with $ClNO_2$ produced from chloride-containing particles. Numerous field-based observational and modeling studies have struggled to accurately simulate ClNO2 production. We developed a new approach to parametrize N_2O_5 uptake and $ClNO_2$ yield that considers the heterogeneity of the aerosol population and applied this in three wintertime environments in Michigan and Maine, USA. Single-particle mass spectrometry and electron microscopy with energy-dispersive X-ray spectroscopy measured single-particle composition and showed that only a fraction of the particulate surface area in each study contained chloride. Our new chemically-resolved surface area parameterization weights N_2O_5 uptake and $ClNO_2$ yield values based on particle type surface area concentrations. This new approach more accurately simulates $ClNO_2$, compared to existing methods that frequently overestimate $ClNO_2$ production. We also use our single-particle measurements to identify sources of particle-phase chloride in the inland environment. We expect that this new approach to simulating heterogeneous reactions will improve modeling of many other heterogeneous reactions that occur at the particle surface.

Early Career Scientist

NO, I am not an early career scientist.

IGAC Activities

CATCH: the Cryosphere and Atmospheric Chemistry