4.065 The Impact of Particle Size, Phase, and Organic Compounds on Interactions between Trace Gases and Particles.

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Abstract:

Heterogeneous reactions between trace gases and atmospheric aerosols impact the oxidative capacity of the atmosphere, regional air quality, and climate. Despite the atmospheric significance of gas/particle interactions, these reactions remain poorly constrained in atmospheric models due to a lack of understanding of how the physico-chemical properties of atmospheric particles impact reaction rates. Using N_2O_5(g) as a benchmark reactive gas, we explore the role of organic coatings and particle size on the reactive uptake of N_2O_5(g). Compared to the reactive uptake of N_2O_5(g) on pure ammonium bisulfate particles, particles composed of mixtures of ammonium bisulfate and organic compounds exhibited highly variable trends in the reactive uptake of N_2O_5(g), which was found to depend on particle phase and the oxygen:carbon ratio (O:C) of the organic material. Organic compounds that significantly inhibited the reactive uptake of N_2O_5(g) were found to lower the diffusivity and/or solubility of N_2O_5(g) in the particle. Inorganic particle composition was found to impact the length that N_2O_5(g) travels within a particle before reacting, referred to as the reacto-diffusive length (l). The reactive uptake of N_2O_5(g) for ammonium sulfates exhibited a size-dependency with uptake coefficients ranging from 0.016 ± 0.005 to 0.036 ± 0.001 as the surface-area weighted particle radius increased from 39 to 127 nm. In contrast, the reactive uptake of N_2O_5(g) on sodium chloride particles was independent of particle size, suggesting that N_2O_5(g) reacts near the particle surface. Differences in the reactivity of the N_2O_5 intermediate, NO_2^+, with water and chloride can explain the dependencies of the reactive uptake of N_2O_5(g) on particle size. These results suggest that parameterizations should factor in the size-resolved composition of ambient aerosols in order to accurately assess the impact of heterogeneous reactions on air quality and climate.