Theme 1. Exposure: Transport and Transformations

Transformation of nanoparticles in the atmosphere

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Reason for research
To understand how the atmosphere modifies nanoparticle properties and how atmospheric processes control the deposition of nanoparticles to ecosystems.

Hypothesis
All emitted nanoparticles will be coated by condensable vapors within hours of emission. Coatings will include inorganics (sulfate, nitrate, and ammonium) and numerous semivolatile organics. Coating will enable particles to act as condensation nuclei for cloud droplets, which in turn will control their deposition to surface ecosystems via rainout. Coating may also alter the surface properties of nanoparticles and thus change their ecosystem impacts.

Objectives
Experimentally test nanoparticle growth via secondary organic aerosol (SOA) condensation and model atmospheric growth rates via both SOA and inorganic (sulfate) condensation. Develop a reactor to coat particles for subsequent testing in other CEINT projects.

Approach
To measure growth via SOA condensation we combined a tandem differential mobility analyzer with a particle-coating system, which includes the generation of nanoparticle aerosol and the production low-vapor pressure organics. We then developed a dynamic mathematical model to describe nanoparticle growth in the atmosphere. The model accounts for the production and subsequent condensation of sulfate, as well as the production and phase partitioning of organics.

Results
Experiments showed that SOA vapors will coat nanoparticles in a flow reactor and cause condensational growth. Predicting growth in the atmosphere is much more subtle, however, because organics form complex mixtures, and background aerosol particles generally contain those rich mixtures. This background of semivolatile organics has very interesting consequences, shown below. In case (a) a nanoparticle is coated with sulfate (red) and a small amount of organics (green) when the organic vapors are added (or formed) with a vapor pressure higher than the vapor pressure of the background organics (but comparable to the vapor pressure of compounds in the coating experiments mentioned above). This is because the background organics serve to suppress the partial pressure of the added organics and thus prevent coating. However, in case (b) the added organics have a lower vapor pressure than the background organics. In this case the added organics coat the nanoparticles and then force additional condensation of organics from the background particles, causing dramatically larger growth.
Implications
These findings have wide implications for nanoparticles in the environment as well as atmospheric aerosol physics in general. The subtle role of semivolatile organics in nanoparticle growth has not previously been explored, and these results show that the ability of organics to coat previously uncoated particles is critically dependent on the relative volatility of the newly formed vapors and the background aerosol. However, in the presence of both organic and inorganic vapors, nanoparticles will grow, and they will typically grow to a sufficient size to nucleate cloud drops (and thus rain out of the atmosphere) in roughly one day.