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Effect of aging on morphology, hygroscopicity, and optical properties of soot aerosols

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Soot from incomplete combustion represents one of the major forms of particulate matter pollution, profoundly impacting human health, air quality, and climate. The direct and indirect radiative effects of soot aerosol depend on particle composition and morphology, which may vary significantly when aerosol is subjected to atmospheric aging. We will present experimental measurements performed in our laboratory to study the effect of internal mixing with atmospheric species on morphology, hygroscopicity, and optical properties of combustion soot. In our experiments, size-classified soot aerosol was exposed to 0.1 - 1000 ppb (part per billion) mixing ratios of sulfuric acid and dicarboxylic organic acids and resulting changes particle morphology and mixing state under dry and humid conditions were characterized through mass-mobility measurements by aerosol particle mass analyzer (APM) and tandem differential mobility analyzer (TDMA). Light absorption and scattering cross-sections for well-characterized fresh and coated soot aerosol were derived using a cavity ring-down spectrometer and an integrating nephelometer in order to assess the effect of atmospheric processing on the radiative properties of atmospheric soot. Internally mixed soot shows significant changes in particle morphology, increasing with the mass fraction of the coating material and relative humidity. Restructuring was the strongest for aggregates coated by sulfuric and glutaric acids whereas succinic acid coating did not result in observable morphology change. Sulfuric acid - coated particles experienced large hygroscopic growth at sub-saturated conditions and activated to cloud droplets at atmospherically relevant supersaturations. Furthermore, coating and subsequent hygroscopic growth considerably altered the optical properties of soot aerosol, increasing light scattering and absorption cross-sections. We found that irreversible restructuring of soot aggregates is a major contributor to the enhancement in optical properties for internally mixed soot. Extrapolation of our results to atmospheric conditions shows that condensation of sulfuric acid is likely a major mechanism of soot aging with profound implications atmospheric soot lifetime and direct and indirect climate forcing.