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Exposure Beyond Mass: High-spatial Resolution Exposure to Source-resolved Atmospheric Particle Number and Chemical Mixing State

**\*Background.\*** Most epidemiological studies address health effects of atmospheric particulate matter (PM) using mass-based measurements as exposure surrogates. These measurements use bulk sampling techniques and therefore are blind to many critical physiochemical properties of individual atmospheric particles. These properties control the deposition of particles in the human lung and likely their toxicity; in addition, they likely have larger spatial variability than PM mass. This study is designed to quantify the spatial variability in number, size, source, and chemical mixing state of individual particles in a populous urban area. We quantified the population exposure to these detailed particle properties and compared them to bulk mass-based exposures.

**\*Methods.\*** We performed mobile sampling using an advanced single-particle mass spectrometer to measure the spatial variability of number concentration of source-resolved 50–1,000 nm particles and particle mixing state in Pittsburgh, Pennsylvania. We built land-use regression models to estimate their spatial patterns and coupled them with demographic data to estimate population exposure.

**\*Results.\*** Particle number concentration had a much larger spatial variability than mass concentration within the city. Freshly emitted particles from traffic and cooking drive the variability in particle number, but mass concentrations are dominated by aged background particles composed of secondary materials. In addition, people exposed to elevated number concentrations of atmospheric particles are also exposed to more externally mixed particles.

**\*Conclusions.\*** Our advanced measurement technique provides a new exposure picture that resolves the large intra-city spatial heterogeneity in traffic and cooking particle number concentrations in the populous urban area. Our results provide a complementary and more detailed perspective compared with bulk measurements of composition. In addition, given the influence of particle mixing state on properties such as particle deposition in the lung, the large spatial gradients of chemical mixing state may significantly influence the health effects of fine PM.