

Combination of dark-field scattering and surface-enhanced Raman spectroscopy for the characterization of submicron individual airborne particles

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Raman microspectrometry (RMS) is a powerful single-particle analytical technique that provides information on the functional groups, molecular species, and mixing states of individual atmospheric aerosol particles. However, drawbacks such as low Raman cross-section, spatial resolution (~1 μm), and optical diffraction limit make it difficult to investigate atmospheric particles in the submicron size range using conventional RMS. Here, we present an advanced RMS technique combining dark-field scattering and surface-enhanced Raman spectroscopy (DF-SERS) for the investigation of atmospheric particles in the submicron size range. Surface-enhanced Raman spectroscopy (SERS) is a useful technique that greatly increases the amount of Raman scattering to characterize trace amounts of analytes. Dark-field scattering facilitates the clear distinction of particles that are below the diffraction limit of optical microscopy (~200 nm). A SERS-active substrate suitable for the analysis of particle form analytes with reproducible hot spots over a large area was fabricated using a sputter coating method. The performance of DF-SERS was evaluated using polystyrene (PS) standard particles in a size range of 60~200 nm. The combined use of dark-field scattering and SERS can provide visual and spectroscopic characterization even for individual particles down to the ~60 nm size. Ambient aerosol particles in a size range of 0.03-1 μm collected on the SERS-active substrate were investigated using DF-SERS. Various organic and inorganic functional groups present in individual particles and their heterogeneous mixing states were successfully investigated, showing that DF-SERS has the potential to provide improved information on submicron atmospheric particles.