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Measuring the pH of Micrometer-Sized Aerosol Particles with using Surface-Enhanced Raman Spectroscopy: A Novel Analytical Approach

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The pH level of atmospheric aerosols plays a crucial role in regulating gas-particle partitioning, phase separation, and heterogeneous reactions. Until now, investigations into the pH of atmospheric aerosols have mainly been conducted using modeling systems such as extended-aerosol inorganic model (E-AIM) or ISOPREPIA-II, or through direct measurements on idealized systems. However, measuring the pH of individual aerosols is essential as it can differ from the pH of the bulk solution and the average pH of atmospheric aerosols, but this has been challenging due to the lack of a suitable analytical technique capable of analyzing small quantities of individual micrometer-sized particles, weighing $\sim 10^{-12}$ g. This study presents a novel analytical technique that uses surface-enhanced Raman spectroscopy (SERS) to investigate the pH of individual aerosol particles, both laboratory-generated and ambient, with sizes in the micrometer range. A highly sensitive and reproducible SERS-active substrate was fabricated through a silver mirror reaction, followed by coating the surface with a self-assembled monolayer of 4-mercaptopyridine (4-MPY) to act as a pH indicator for the collection and analysis of individual particles. Aerosol particles, similar in size to ambient aerosols, were nebulized on the substrate, and measurements were carried out in an in-situ system that stabilized the relative humidity to maintain the water content of individual particles. The pH of individual particles was determined based on the ratio of two pH-sensitive Raman peaks of the 4-MPY molecule at 1580 and 1620 cm^{-1}. This technique successfully discriminated aerosol particles in the pH range of 1-7, demonstrating its potential to examine ambient aerosol particle pH, as atmospheric aerosols are predominantly acidic. The feasibility of this method was validated using standard ammonium sulfate aerosols, a major component of secondary inorganic aerosols. In addition, both the pH and chemical composition of ambient aerosols were investigated using SERS and SEM/EDX for the same individual particles. Measuring the pH of individual aerosol particles is crucial in gaining a better understanding of the impacts of atmospheric aerosols on climate change and human health.