**Simultaneous Characterization of n-Alcohols (C=1-4) at and Below Aerosol Surfaces**

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**Abstract**

Atmospheric aerosol particles are abundant and ubiquitous in both indoor and outdoor environments. Such species have an understudied effect on the environment and human wellbeing due to their unique interfacial properties. While it is widely accepted that the chemical and physical evolution of aerosol particles in the atmosphere is closely connected to interface-specific phenomena, this topic has yet to be investigated by methods which are noninvasive and nondestructive. In this study we present the novel technique of vibrational sum frequency scattering (VSFS) spectroscopy as an interface-specific, high-performance method for the in-situ investigation of the gas/aerosol particle interface, as well as its underlying bulk phase through simultaneous hyper-Raman scattering (HRS) spectroscopy. We use aqueous alcohol aerosols to demonstrate this technique's ability to separate the vibrational phenomena which take place at the gas/particle interface from those in the particle's bulk phase. Observing aerosol particles containing methanol, ethanol, 1-propanol, and 1-butanol through VSFS provides interfacial molecular characterization. Additionally, we demonstrate interfacial differences between aerosol particles and their planar analogue. The sensitivity of this technique to probe the interface of laboratory-generated aerosol particles with few-particle density at standard conditions validates VSFS as an analytical technique for environmentally relevant aerosols.