

***In Situ* Chemical Structure Detection of Organic Species at Aerosol Particle Surfaces
and in Bulk Phase**

Yuqin Qian, Hui Wang, Jesse B. Brown, Zhi-Chao Huang-Fu, Tong Zhang, and Yi Rao*

Department of Chemistry and Biochemistry, Utah State University, Logan, UT 84322

Abstract

The unique chemical and physical properties of aerosol particles is important to scientists, engineers, and medical professionals due to their profound effects on air quality, human health, and environmental and atmospheric chemistry. The growth processes of naturally and anthropogenically generated aerosols is inherently connected with the interactions of gas phase species and those at the surface of aerosol particles, as well as reactions within the bulk phase of the particle. Nonetheless, direct observations of chemical structures at the gas/aerosol particle interface are deficient. In this work we demonstrate in-situ surface-specific vibrational sum frequency scattering (VSFS) spectroscopy to directly identify chemical structures of short-chained carboxylic acid species at the surface of aerosol particles. Additionally, this setup allows the simultaneous probing of the particle's bulk phase through hyper-Raman scattering (HRS) spectroscopy. Polarization-resolved VSFS spectra of propionic acid at the aerosol surface were examined, as well as vibrational spectra from this acid in the particle bulk. Furthermore, the adsorption free energy of propionic acid to the aerosol particle surface was compared to that of the planar liquid surface, showing a lower value for the particle surface. These results demonstrate that the traditional analogy between the curved and planar surface may be incomplete. This novel technique offers an approach to the study of surface-active species in aerosols, as well as their aging processes in-situ, and chemical analysis of indoor and outdoor viral aerosol particles.