***Effects of Relative Humidity and Salt-Out on Interfacial Behaviors of Molecules on Aerosols***

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The interfacial roles of aerosols in chemical processes of the atmosphere have long been recognized. We here developed an interface-sensitive Second Harmonic Scattering (SRS), to directly observe molecular behaviors at laboratory-generated aerosol interfaces. We, for the first time, successfully probed interfacial population of organic molecules on aerosols floating in the gas. We found that the surface driving force for the curved aerosol surfaces is less than that for the planar air/water interface. We attributed the weaker driving force at aerosol surfaces to the significant amount of <100 nm aerosol particles, which have smaller surface tensions as compared to the larger curved particles. These results challenge the long-standing wisdom that aerosol surfaces facilitate stronger surface binding of molecules than the air-water interface. Our findings would open a new avenue for the studies of chemical and physical properties of aerosols. We also investigated the effect of Salt and relative humidity on surface adsorption of aerosols. For the salting-out effect, we measured SHG of interfacial molecules, DIA 4, in different salt solutions, including NaCl, Na2SO4, NH4NO3, and (NH4)2SO4 to form aerosol particles. The surface adsorption abilities of DIA 4 of aerosols from the four salts are similar. On the other hand, it is surprising that surface populations are different, exhibiting an increasing tendency from Na2SO4, NaCl, (NH4)2SO4, to NH4NO3. Measurements of SHG were also made for surface adsorption of Dia 4 of aerosols under different relative humidity. Relative humidity influenced surface adsorption of Dia 4 at the aerosol surface. At the relative humidity of 85% surface adsorption of Dia 4 of aerosols is stronger than that at the relative humidity of 45%.