2.145 First steps to uncovering molecular level interactions of organic aerosol and cloud droplets from direct observation using synchrotron radiation..

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Abstract:

Advances in aerosol measurement techniques and model formulation have highlighted key features still lacking in our fundamental understanding and process description of especially the organic aerosol fraction. Discrepancies between experimental setups and between models and experimental results are evident in studies of new particle formation, aerosol growth and cloud activation, and atmospheric scale cloud effects. As a result, uncertainties in aerosol climate effects remain almost unchanged for decades.
We explore the potential for emerging experimental methods using high-brilliance synchrotron radiation (SR) to shed light on molecular-level interactions between organic aerosol and atmospheric water. Using highly surface sensitive and chemically specific Xray photoelectron spectroscopy (XPS) in combination with SR, we directly observe shifted protonation equilibria of organics with carboxylic acid functionalities in the aqueous surface. We also found isomer effects in the surface behavior of aqueous alcohols and used directly observed concentration-dependent surface compositions to evaluate a novel statistical mechanics based model for surface active organics. Using XPS on free-frying salt clusters with varying water content, we observed structural properties and solvation of sub-2 nm particles, including relative depth profiles and concentration dependence of ion solvation and cluster-size dependent phase transition of the salt crystal lattice structure.
Current efforts focus on enhancing the immediate atmospheric relevance of experimental design, including moving towards ambient pressures and single particle in-situ studies. The emergence of fourth generation SR facilities like MAX IV Laboratory in Sweden brings great promise in this respect. We are part of the consortium commissioning the Finnish-Estonian Beamline for Atmospheric and Materials Science (FinEstBeAMS). With a wide photon energy range, high brilliance and resolving power, and three complementary end-stations, the beamline enables studies of a wide range of systems, conditions and properties. We are furthermore developing mobile instrumentation to specifically enable atmospherically directed research at several beamlines at MAX IV.