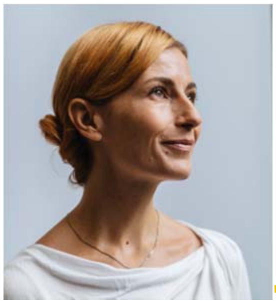
Aqueous organic surfaces: from synchrotron experiments to climate models



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We study multiphase and multiscale atmospheric processes and their impact on air pollution and climate. Our current work combines synchrotron radiation based spectroscopy and imaging, artificial intelligence, and classical thermodynamics.

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Submicron aerosol particles and cloud droplets have very large surface area relative to their volume, but are challenging to characterize using typical bulk-sensitive methods due to the very small overall amounts of mass confined in the surface. Surface sensitive analytical techniques have recently revealed surface chemical compositions of atmospherically relevant systems, which are very different from the associated condensed bulk phase. While expected from classical adsorption theory, significant challenges remain in obtaining quantitative closure between surface composition obtained from state-of-the-art thermodynamic surface models and surface sensitive experiments. These discrepancies have guided both our experimental design and development of novel surface thermodynamic frameworks.

We have probed surface composition of various aqueous organics in atmospherically relevant model mixtures using different synchrotron radiation excited techniques, in particular x-ray photoelectron spectroscopy (XPS). Experiments were performed under a range conditions using specialized end-station environments at several synchrotron facilities, including the Finnish-Estonian Beamline for Atmospheric and Materials Science (FinEstBeAMS), HIPPIE, and SPECIES at MAX IV, and beamlines at UVSOR and BESSY II. Results are in line with earlier work from MAX II (beamline I411)[1-3], but also reveal several new features. Comparing experimental results to predictions from both classical and statistical thermodynamics based adsorption frameworks, including both Gibbsian models and a novel monolayer adsorption model, reveal challenges relating both to the theoretical characterization of non-ideal solution phenomena, and the interpretation of experimental data[4-6].

The description of aqueous surfaces has important implications for predictions of atmospheric cloud droplet formation and droplet radiative and chemical properties through several processes governed by the surface, including highly composition sensitive surface tension reduction[7].

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