NOVEL INSIGHTS TO AQUEOUS ORGANIC SURFACES AND CLUES TO UNRAVELING THEIR ATMOSPHERIC IMPACTS

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Tiny aerosol particles, with dimensions between a few micro-meters and just a few nanometers, are everywhere in the atmosphere, from the most polluted mega-cities to the pristine Arctic air. These aerosol particles have profound impacts in the atmosphere. In Europe alone, air pollution and predominantly aerosols currently causes over 350.000 premature deaths per year. Aerosols also play a key role in regulating Earth's climate via their interactions with the global radiation balance and impacts on cloud formation [1]. Aerosol interactions with atmospheric water are central to each of these effects. A significant fraction of atmospheric aerosol mass is organic and often display surface activity in aqueous mixtures. My previous work on aerosol cloud formation thermodynamics has demonstrated a consistent inability to reconcile experimental and bulk theoretical results, which strongly suggests that the surface composition and resulting surface tension of aqueous organic aerosols are significantly different from the bulk aerosol phase [2]. We then demonstrated the potential for such differences to impact aerosol-climate interaction all the way to the global scale [3].

Until very recently, there has been no experimental techniques by which to verify this by direct observation. Development of the liquid micro-jet technique has in the last decade made it possible to use highly surface sensitive and chemically selective synchrotron-radiation based Xray photoelectron spectroscopy (XPS) for studying systems with finite vapor pressures, and in particular for atmospherically relevant aqueous and organic mixtures. We recently successfully used XPS to make several surprising discoveries of surface-specific organic speciation, chemical reactivity, and aqueous acidity for such systems [4]. None of these surface properties are currently considered in our understanding or predictions of aerosol effects in the atmosphere. Here, I present a framework to begin including highly surface-specific chemical properties gained with XPS in aerosol thermodynamics and large-scale processes, with potentially dramatic atmospheric impacts. For example, surface contributions could contribute to unravel a long-standing underestimation by factor of 5 of atmospheric organic aerosol formation, the so-called "missing SOA mystery".

[1] Intergovernmental Panel on Climate Change. Climate Change 2013: The Physical Science Basis. *Cambridge University Press*, New York, USA.

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[3] Prisle, N. L., Asmi, A., et al. (2012). *Geophys. Res. Lett.*, 39:L05802.

[4] Prisle, N. L., Ottosson, N., et al. (2012). *Atmos. Chem. Phys.*, 12:12227–12242.