Geophysical Research Abstracts
Vol. 18, EGU2016-10890, 2016
EGU General Assembly 2016

Water interactions with condensed organic phases: a combined experimental and theoretical study of molecular-level processes

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Water uptake on aerosol particles modifies their chemistry and microphysics with important implications for air quality and climate. A large fraction of the atmospheric aerosol consists of organic aerosol particles or inorganic particles with condensed organic components. Here, we combine laboratory studies using the environmental molecular beam (EMB) method1 with molecular dynamics (MD) simulations to characterize water interactions with organic surfaces in detail. The over-arching aim is to characterize the mechanisms that govern water uptake, in order to guide the development of physics-based models to be used in atmospheric modelling.

The EMB method enables molecular level studies of interactions between gases and volatile surfaces at near ambient pressure,1 and the technique may provide information about collision dynamics, surface and bulk accommodation, desorption and diffusion kinetics. Molecular dynamics simulations provide complementary information about the collision dynamics and initial interactions between gas molecules and the condensed phase.

Here, we focus on water interactions with condensed alcohol phases that serve as highly simplified proxies for systems in the environment. Gas-surface collisions are in general found to be highly inelastic and result in efficient surface accommodation of water molecules. As a consequence, surface accommodation of water can be safely assumed to be close to unity under typical ambient conditions. Bulk accommodation is inefficient on solid alcohol and the condensed materials appear to produce hydrophobic surface structures, with limited opportunities for adsorbed water to form hydrogen bonds with surface molecules. Accommodation is significantly more efficient on the dynamic liquid alcohol surfaces. The results for n-butanol (BuOH) are particularly intriguing where substantial changes in water accommodation taking place over a 10 K interval below and above the BuOH melting point.2 The governing mechanisms for the observed water accommodation are discussed based on the combined EMB and MD results. The studies illustrate that the detailed surface properties of the condensed organic phase may substantially modify water uptake, with potential implications for the properties and action of aerosols and clouds in the Earth system.

References:
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