Bridging experiments and molecular simulations to elucidate heterogeneous ice nucleation

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Heterogeneous ice nucleation is the primary pathway for ice formation in the atmosphere. Different types of particles such as pollen, bacteria, and mineral dusts influence ice nucleation. In some cases, these particles allow ice to form at temperatures as high as -3^oC. In comparison, homogeneous ice nucleation occurs at -40^oC. Currently, the understanding of the interplay between surface's chemical and physical properties and its ability to nucleate ice is lacking. There is no framework to predict the ice nucleating ability of a surface based on it's characteristics. The primary challenge is the inability to access the relevant length and time scales. Nucleation occurs in nano-to-micro second timescales and lengthscales of few hundred-to-thousand molecules. These are difficult to probe in experiments. In principle, these are ideal scales for molecular simulations, however, the challenge comes from the fact that ice nucleation is a rare event – i.e., an event which occurs at frequencies lower than the simulation sampling time.

Using advanced sampling techniques combined with molecular dynamics simulations, we have evaluated the role of various surface properties on ice nucleation. We studied three types of surfaces – kaolinite-based, silver iodide-like and mica surfaces. Capitalizing on the power of molecular simulations we have carefully probed the effects of specific surface properties such as hydrogen bonding abilities, lattice spacing, and surface charge distribution on the propensity to observe ice nucleation. We find that certain characteristics of the liquid water structure – related to the orientations of water molecules near the surface – are good indicators of possible ice nucleation.[1,2] We are expanding our understanding to correlate these observations with experimental findings. To this end, we focus on mica surfaces. Mica surfaces can be cleaved to be atomistically smooth in experiments. This makes them an ideal choice for a collaborative experimental and simulation investigation since in simulations the surfaces are usually made atomically smooth. We study the effects of surface ion, charge distribution and lattice spacing on ice nucleation. Our simulations indicate that the interfacial structure of water near the mica surfaces provides signature characteristics to predict the ice nucleating propensity. In experiments, we have used Fourier Transform Infrared Spectroscopy to study the liquid water structure near the mica surfaces.[3,4] Through this combined approach, we evaluate the role of surface chemistry on ice nucleation. In this talk, we will present these results and discuss how we bridge experimental and simulations findings.

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[2] Glatz, B. and Sarupria, S, "The surface charge distribution affects the ice nucleating efficiency of silver iodide", Journal of Chemical Physics 145, 211924, 2016

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[4] Cantrell, W. and G. Ewing. "Attenuated (but not Total) Internal Reflection FTIR spectroscopy of thin films". Applied Spectroscopy, 56, 665-669, 2002.