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Polycyclic Aromatic Hydrocarbons on Ice

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Ice has significant effects on the climate and biogeochemical systems and plays a role in reactions known to contribute to ozone depletion and atmospheric pollution. Long range transport of hazardous pollutants, such as polycyclic aromatic hydrocarbons (PAHs), on ice contributes to the disruption of these biogeochemical systems. PAHs and their derivatives from photolysis are known to be carcinogenic to humans and hazardous to plants and animals. Despite the global impact, the adsorption of PAHs and their derivatives on ice surfaces and the mechanism for photolysis have yet to be determined. To observe the absorption of PAHs on ice surfaces, a surface-selective technique with molecular specificity is essential. Moreover, to study the photolysis of PAHs on ice surfaces in real time ultrafast time resolved measurements are required. Time resolved sum frequency generation spectroscopy (SFG) is a surface selective technique that probes reactions on sub-picosecond timescales by monitoring vibrations of molecules especially at an interface. Therefore, measuring PAHs on ice surfaces with SFG spectroscopy would provide a molecular scale understanding of the adsorption of PAHs on ice surfaces and the photolysis mechanism. As a first step in unraveling the mechanism, we probe the adsorption of naphthalene and anthracene on the surface of well-defined single crystalline ice, using molecular vibrations intrinsic to both the PAHs and ice. Determining the adsorption of PAHs on ice surfaces and the role of ice in their subsequent photochemical fate can resolve which molecules have a higher propensity for the ice surface and how their photo reactivity is affected. Such insights will aid in understanding both long range transport and surface reactivity of contaminants. In this way, better models to predict the disruption of biogeochemical cycles can be developed.

Significance statement

Ice acts like a sink for organic pollutants and can serve as catalyst and reaction site. Specifically, upon adsorption on ice, PAHs undergo significant red shifts in their absorption spectra, overlapping with the solar output. We aim to elucidate adsorption and photochemical reactions of PAHs on ice surfaces.

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