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Adsorption and dissociation of acidic trace gases on ice surfaces - caught in the act with core level spectroscopy

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Chemistry and physical processes in Earth's ice and snow cover can change the composition of the atmosphere and the contaminant content of the cryosphere. They have thus direct impacts on geochemical cycles and the climate system. Our ability to predict the fate of chemicals in snow or air masses in exchange with the cryosphere on a regional scale or to model those in snow chemistry models is currently hampered by our limited understanding of the underlying mechanisms on a molecular level.

So far, direct experimental observations under environmentally relevant conditions of the ice surface and of the adsorption of trace gases to it are very limited. The unique approach of this study is to combine two surface sensitive spectroscopic methods to directly probe the hydrogen-bonding network at the ice surface ($\sim$1 nm depth) and the concentration, depth profile ($\sim$1 to 10 nm), and dissociation degree of the dopant. We present first core-electron photoemission (XPS) and partial electron yield X-ray absorption (NEXAFS) measurements of formic acid adsorbed to ice at 240 K. The analysis of oxygen NEXAFS spectra reveals information on changes in the hydrogen-bonding network of the ice surface upon adsorption of formic acid. Depth profiles based on XPS measurements indicate that the adsorbed acid stays at the ice surface. Furthermore we obtained a preliminary estimation of the degree of formic acid dissociation at the ice surface.

Results are compared to earlier core-electron studies of several trace gases adsorbed to ice at 240 K and compared to results from more traditional method to and snow to reveal fundamental aspects of the ice surface and how it interacts with dopants. Even with the focus on adsorption of acidic trace gases to ice, results of this study will thus be of high relevance also for other chemical processes in ice and snow. This is of interest not only in environmental science but also in material science, cryobiology, and astrophysics.