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A spectroscopic look at the ice – air interface under the effect of adsorbing gases

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Chemical processes on ice in snow and cirrus clouds are important for atmospheric chemistry, biogeochemical cycling of trace constituents and archiving of trace species in ice. It has been suspected since long that trace contaminants affect the thickness of the quasi-liquid layer at the ice - air interface, which is at the heart of the interactions between snowpacks or cirrus clouds and the surrounding air. Nitric acid is highly soluble in water and has a strong affinity to ice. It has been suggested to be a source of nitrogen oxides through photolysis. Hydrogen chloride is a key species in stratospheric ozone depletion through its reaction with chlorine nitrate on ice or within liquid particles. Acetone is a ubiquitous small oxygenated organic gas that provides a source of HOx in snow or in the upper troposphere. All three species have been extensively studied with respect to their partitioning behavior and uptake kinetics to ice. However, the majority of these investigations have derived uptake to ice indirectly through the loss of the species of interest from the gas phase, and important issues remain with the real processes at the ice surface to explain the contrasting behavior of these species. Here we show the results of X-ray photoelectron spectroscopy (XPS) experiments performed at water vapor pressures in equilibrium with ice in the temperature range 220 to 250 K, which allow to obtain surface chemical composition and near surface depth profiles of ice exposed to trace gases at atmospherically relevant pressures. Electron yield near edge X-ray absorption spectroscopy (NEXAFS) further allows probing the local hydrogen bonding environment at the surface, i.e., the quasi-liquid layer (QLL).