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Depth Profiles of Solutes at the air-ice Interface

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The pre-melting at the surface of ice crystals in surface snow or ice clouds has been proposed to explain a number of large-scale environmental effects ranging from electrification of thunder clouds and the scavenging of atmospheric trace gases to the flow of glaciers¹. There is now general agreement on the appearance of this quasi-liquid layer (QLL) when temperatures approach the melting point and on its enhancement in presence of impurities². In particular, McNeill showed that the extended quasi-liquid layer increases the solubility of trace gases leading to a larger capacity for scavenging from the gas-phase and that it accelerates chemical reaction rates of atmospheric importance³. While this is an eminent example of how anthropogenic emissions can alter the properties of clouds and surface snow, the precise on-set temperature of this impurity induced QLL, its properties and extend remain controversial.

Here, we present experimental results directly showing how deep a number of formic acid, nitric acid, and hydrochloric acid enter the upper few nanometre of the air-ice interface and to which extend their presence is accompanied by the formation of a QLL. These findings using a combination of near ambient pressure X-ray photoelectron spectroscopy (XPS) ^{4,5} and partial electron yield near edge X-ray absorption fine structure (NEXAFS) ^{4,5} spectroscopy reveal that the ability of these acids to induce a QLL is a function of temperature and scales with room temperature solubility. Direct comparison of the thickness of the QLL as interfered by the NEXAFS measurements with the depth of the acids within the QLL indicate that the QLL is not a homogeneous layer. Rather, we propose that the acids induce solvation shells in their vicinity. The observed extend of QLL, or hydration shell formation, is then a balance of the individual acid's water demand to satisfy their hydration needs and the liquid-like properties of the ice surface which is a function of temperature and impurity content. Further, the dissociation degree of acids in the QLL is discussed.

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Further information:

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Significance statement

Overall, this study shows how trace gases can alter the properties of ice and snow on a molecular level in the Anthropocene with implications for multiphase chemistry, nucleation, and physical properties of surface snow and ice clouds.

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