Geophysical Research Abstracts
Vol. 21, EGU2019-8899, 2019
EGU General Assembly 2019

Hydrogen Bonding at the Ice Surface

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We elucidate the structure of interfacial water molecules at the surface of solid ice, using surface-specific vibrational spectroscopy of interfacial water molecules. While in the bulk of ice, each water molecule is fourfold coordinated, both accepting and donating two hydrogen bonds, the hydrogen-bonded network is abruptly interrupted at the surface. The resulting under-coordination of ice molecules at the interface determines many of the properties of the ice surface. Using single-crystal ice, under well-defined conditions, we study the temperature dependence of the outermost surface monolayer of water molecules. We find an excess of hydrogen bonds at the ice-vapor interface around 200 K, due to a competition between entropic and enthalpic contributions to the free energy. At higher temperatures, surface melting of ice is found to occur in a bilayer-by-bilayer fashion. Finally, we show that the temperature-dependent interfacial molecular structure correlates with the temperature dependent macroscopic friction coefficient of ice.