Geophysical Research Abstracts
Vol. 14, EGU2012-9447, 2012
EGU General Assembly 2012

Inefficient formation of ice via heterogeneous nucleation at temperatures below 200 K

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Nucleation of ice on aerosol particles is an important route to the formation of cirrus and other high altitude clouds in the atmosphere. Here we investigate heterogeneous ice nucleation via deposition mode freezing on both hydrophobic and hydrophilic surfaces using environmental molecular beam experiments and molecular dynamics simulations. We observe that nucleation of ice on a bare graphite crystal becomes increasingly inefficient as the surface temperature decreases from 200 to 155 K. The graphite has a hydrophobic character and water does not wet the surface efficiently in this temperature range. Adsorption of a monolayer of methanol on the graphite surface changes it from hydrophobic to highly hydrophilic. The methanol molecules provide sites for efficient hydrogen-bonding of water molecules, which stabilizes water on the surface compared to the bare graphite. Ice nucleation on the hydrophilic surface takes place at a lower supersaturation than on the hydrophobic surface, and the adsorbate thus influences the absolute nucleation rate at a given temperature. However, the supersaturation required for nucleation increases rapidly with decreasing temperature in the range 175-190 K, and the overall trend with temperature is similar for the bare and methanol-covered surface. Adsorption of a butanol monolayer results in an ice nucleation efficiency intermediate between the other systems. Butanol forms a highly stable solid layer on graphite. Water does not appear to wet the butanol layer efficiently and the water stability on the surface is lower than on the methanol layer. Again, the trend with temperature is similar to the other investigated systems. Thus, while the hydrophilicity of the different surfaces influences the absolute nucleation rate, the overall trend with temperature remains similar. The combination of the present investigations of carbon-based hydrophobic and hydrophilic systems with existing literature provides us with a sufficient data set to allow us to generalize the behavior of the deposition freezing process at low temperatures. Although the substrate plays a role, the overall trends with temperature are similar for different surfaces and therefore the explanation for the observed inefficient nucleation must be related to the inherent properties of water at low temperature. The importance of the results for cloud formation processes in the atmospheres on Earth and Mars are discussed.