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Towards a molecular-level understanding of heterogeneous ice nucleation by direct surface deposition of water vapor

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Ice nucleation research is currently booming and much of the attention is motivated by the need to improve the description of aerosol and cloud processes in the atmosphere. The formation of clouds has important effects on the water cycle on Earth and on the radiation budget of the atmosphere. The formation of liquid cloud droplets is comparatively well described by existing theory, while the formation of ice particles remains poorly understood. This is a major concern since it introduces uncertainties in our ability to model and project changes in climate.

In the atmosphere water droplets tend to remain in a super-cooled state far below the melting point of water with temperatures of about 235 K required to homogeneously freeze pure cloud droplets. Alternatively, ice formation may be enhanced by heterogeneous nucleation that may occur, either when supercooled liquid water contacts an insoluble aerosol particle and subsequently freezes, or when water vapor is directly deposited as ice onto a particle surface.

Here we focus on the latter mechanism that is commonly referred to as deposition freezing. This ice nucleation mode primarily contributes to nucleation at temperatures below approximately 243 K, and is more important than other heterogeneous ice nucleation modes at temperatures below 235 K. Deposition freezing is thus of particular relevance for clouds in the upper troposphere. Similar to other heterogeneous ice nucleation modes, deposition freezing remains poorly understood, and parameterizations currently used in cloud models are mainly based on empirical data from laboratory and field studies.

Although we know that several different types of solid material may act as deposition freezing ice nuclei, we do not yet have a theoretical description that can be used to predict the outcome. Defects in the substrate, the type of bond that the adsorbing water molecules form with the substrate, and the type of atoms exposed on the substrate all appear to affect heterogeneous nucleation.

This paper takes a molecular-level perspective and surface science-based approach to investigating deposition freezing phenomena. We summarize the current understanding of the ice formation process based on recent results obtained with several methods, including environmental molecular beam experiments, molecular dynamics simulations and kinetic modelling. Existing models are critically reviewed and outstanding questions are discussed.

Significance statement

This paper will review the current molecular-level understanding of heterogeneous ice nucleation in the deposition freezing mode, based on recent experiments and theory using methods adopted from the fields of surface and aerosol science. Existing models are critically reviewed and outstanding questions are highlighted and discussed.

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