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Deposition Nucleation or Pore Condensation and Freezing? The role of pores on ice nucleation

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Ice crystals in clouds play an important role in initiating precipitation and thus cloud lifetime which acts to moderate the Earth's radiative balance. As such, understanding the mechanisms responsible for ice formation is necessary to quantify the impacts of anthropogenic climate change. Pore condensation and freezing (PCF), is a proposed mechanism for ice formation below water saturation. During PCF, water condenses in pores or surface imperfections (herein referred to as pores) below water saturation, as predicted by the inverse-Kelvin effect. Thus, the ability of a pore to take up water strongly depends on the pore width and the contact angle of the pore wall. Once liquid water is present, ice can nucleate heterogeneously if an ice active site exists on the pore surface, or homogeneously if the temperature is below 235 K. However, the pore width must also be capable of accommodating the critical ice embryo for ice nucleation to occur. Therefore, the PCF mechanism is limited to particles with a critical pore size narrow enough to fill with water and wide enough to accommodate a critical ice embryo for a given temperature.
To investigate the PCF mechanism, spherical mesoporous silica particles with well-defined pore diameters of 3 to 4 nm are evaluated for their ice nucleation ability in the Zurich Ice Nucleation Chamber (ZINC). Within ZINC, the particles are exposed to temperatures ranging between 223 and 238 K and varying supersaturations with respect to ice. To further test the applicability of the PCF mechanism, the particles are functionalized with methyl and hydroxyl groups to alter the contact angle of the particles. We find that the contact angle of the particles helps determine the relative humidity at which pore condensation occurs and subsequently, ice nucleation. These results, combined with experiments performed with nonporous spherical silica, strongly support the proposed PCF mechanism.

Significance statement

Ice nucleation below water saturation is conventionally viewed as the direct transition from water vapor to ice. However, our results suggest that supercooled liquid water contained in pores and surface imperfections is responsible for ice formation below water saturation.

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