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Inhibition of ice crystallisation in highly viscous aqueous organic acid droplets

B. J. Murray
School of Chemistry, Woodhouse Lane, University of Leeds, Leeds LS2 9JT, UK

Abstract. Homogeneous nucleation of ice within aqueous solution droplets and their subsequent crystallisation is thought to play a significant role in upper tropospheric ice cloud formation. It is normally assumed that homogeneous nucleation will take place at a threshold supersaturation, irrespective of the identity of the solute, and that rapid growth of ice particles will follow immediately after nucleation. However, it is shown here through laboratory experiments that droplets may not readily freeze in the very cold tropical tropopause layer (TTL, typical temperatures of 186–200 K). In these experiments ice crystal growth in citric acid solution droplets did not occur when ice nucleated below 197±6 K. Citric acid, 2-hydroxypropane-1,2,3-tricarboxyllic acid, is a molecule with similar functionality to oxygenated organic compounds which are ubiquitous in atmospheric aerosol. It is therefore thought to be a sensible proxy for atmospheric organic material. Evidence is presented that suggests citric acid solution droplets become ultra-viscous and form glassy solids under atmospherically relevant conditions. Diffusion of liquid water molecules to ice nuclei is expected to be very slow in ultra-viscous solution droplets and nucleation is negligible in glassy droplets; this most likely provides an explanation for the experimentally observed inhibition of ice crystallisation. The implications of ultra-viscous and glassy solution droplets for ice cloud formation and supersaturations in the TTL are discussed.

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