Formation of high-altitude ice clouds: ice nucleation or freezing?

Anatoli Bogdan[1,2] and Mario J. Molina[3]

[1] Laboratory of Polymer Chemistry, Department of Chemistry, University of Helsinki, Finland
[2] Department of Physical Sciences, University of Helsinki, Finland.
[3] Department of Chemistry and Biochemistry, University of California, San Diego, CA, USA

Upper tropospheric (UT) ice cirrus and type II ice PSCs are believed to be formed by freezing aqueous aerosol drops which contain up to 25–30 wt % of H_2SO_4 and HNO_3. There are also reports that in UT aqueous drops, organic component may reach up to 50 wt %. Currently, atmospheric scientific community dealing with high-altitude ice clouds focuses only on the initial step of freezing, namely, on ice nucleation. How freezing proceeds after ice nucleation is not considered, although it is the freezing process itself which governs the phase state and surface properties of resulting cloud particles. Such limited consideration results in the lack of understanding of freezing process and, consequently, is a reason that some important problems of the formation and microphysics of UT cirrus and PSCs and the impact of these clouds on climate remain unsolved for decades. One of the important problems is whether freezing atmospheric aqueous drops produce completely solid ice particles, as is generally believed, or mixed-phased cloud particles in which an ice core is coated with a freeze-concentrated solution (FCS). About freeze-induced phase separation into pure ice and FCS[1] and that mixed-phased cloud particles can be formed in UT[2-5] and polar stratosphere[6] have been reported some time ago. Unfortunately, atmospheric scientists dealing with UT cirrus and PSCs ignore these works that can stem from the fact that they cannot still comprehend and, consequently, accept the well-known fact of freeze-induced phase separation which occurs during the freezing of aqueous solutions, including atmospheric aqueous drops. The goal of this presentation is to present persuasive experimental results, including the visual demonstration of freezing process[7], which would convince the atmospheric scientific community of the freeze-induced phase separation (Figure 1) and, consequently, of the formation of mixed-phase cloud particles at very beginning of UT cirrus and type II ice PSC development (Figure 1b). The knowledge of the phase state and surface properties of cloud particles (cloud microphysics) is important because they govern the rate of heterogeneous reactions destructing stratospheric and UT ozone and radiative properties of UT cirrus (absorption, reflection, and scattering of solar and terrestrial radiation). The FCS coating around UT cirrus particles strongly reduces the uptake of water vapor - the dominant greenhouse gas in UT - and, consequently, is responsible for the accumulation and persistence of elevated UT moisture.[3,4]

References:

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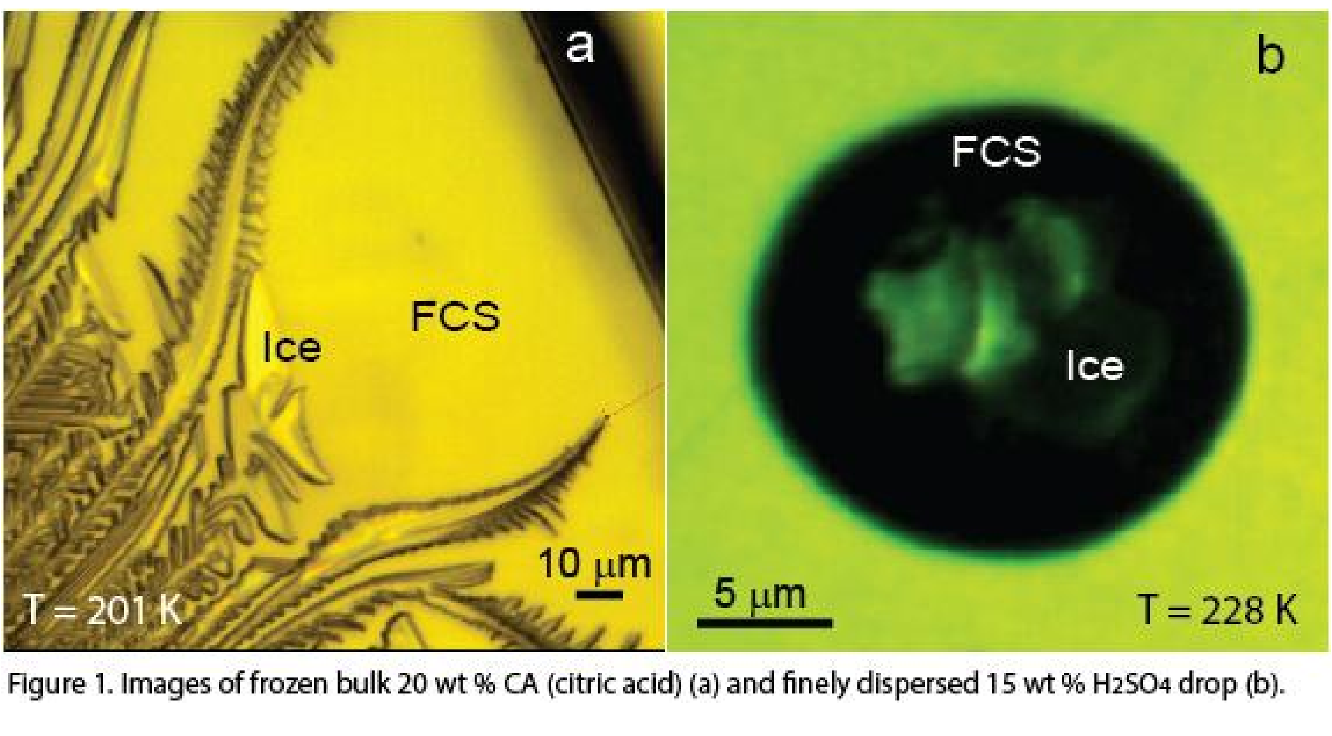

Figure 1. Images of frozen bulk 20 wt % CA (citric acid) (a) and finely dispersed 15 wt % H_2SO_4 drop (b).