LETTER

Reply to Bogdan et al.: "Cubic ice" in cirrus clouds under dry and wet conditions

Bogdan et al. (1) discuss the relevance of surface roughness of vapor-deposited so-called "ice I_c" discovered in ref. 2 for ice crystals formed at low temperatures in cirrus clouds. The authors emphasize that for ice nucleation from solute aerosol droplets, the initial ice crystal is coated by residual freeze-concentrated solutions, and they provide images from laboratory freezing experiments on large H_2SO_4 and NH_4-sulfate solution droplets.

Although cirrus clouds are likely to contain ice formed via freezing of concentrated solutions as discussed in refs. 20, 55, 58, and 59 from ref. 2, at least initially leading to micrometer-sized nearly spherical "droxtals," there is multiple and compelling evidence that this is by far not the only shape of ice particles in cirrus clouds. It should be noted that, in contrast to Bogdan et al. (1), we did not limit our discussion to the incipient stage of ice formation (see refs. 30, 34, 63–66 from ref. 2). The fact that cirrus clouds contain faceted ice crystals is obvious from halo observations but is also supported by satellite reflectance measurements (3) by a direct collection using replicas, in-flight imaging, and light scattering techniques, as summarized in ref. 65 from ref. 2. Thus, uniaxial faceted ice crystals and bullet rosettes are a frequent encounter in cirrus clouds, and it is in this context that we draw our attention to the observation of the kinks in the prismatic facets of crystals grown within the temperature range where we expect "ice I_c" to be formed. These kinks develop on rather mature several-micrometer large prismatic crystals of "ice I_c" of trigonal (pseudohexagonal) symmetry, whereas the incipient ice is more isometric in shape (2). Bullet rosettes may emerge from a cubic nucleus and may also have kinky prismatic facets (see ref. 35 from ref. 2).

Moreover, the still frequently encountered assumption that (acid) aqueous sulfate aerosols are the primary agent for ice nucleation in cirrus clouds has become questionable in recent years. Natural terragenic, biological, and strong anthropogenic ground sources are likely to have a significant impact on the upper troposphere aerosol population (4); the importance of heterogeneous ice nuclei (e.g., mineral dust, soot, organics) on homogenous freezing has been pointed out repeatedly, and the importance of the interplay of ice homogeneous and heterogeneous ice nucleation, diffusional ice growth, and sedimentation of larger ice crystals was highlighted recently in a detailed modeling study (5). As a likely consequence, there is a multitude of ice crystals of different shapes developing in cirrus clouds. In other words, what in ref. 1 has been evoked as "dry" atmospheric surfaces is not the exotic item it may appear to be at first sight.

Undoubtedly, more research is needed to link modeling efforts, laboratory experiments, and air- and space-borne observations. The picture emerging is likely to be complex, multifaceted, and time dependent; the purpose of our contribution (2) was to show that this complexity is further enhanced by the presence of "ice I_c" with its different and constantly changing physical and morphological properties.

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