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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Scanning electron microscopy and molecular dynamics of surfaces of growing and ablating hexagonal ice crystals" *by* W. C. Pfalzgraff et al.

Anonymous Referee #3

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I do not recommend this paper for publication in its current form and I question the relevance of the laboratory results to the real atmosphere for several reasons, the main one being the absence of trans-prismatic strands being observed with in situ crystals. The likely reasons for this lack of observation follows. [I agree with another reviewer: the reference to "Figures 3e-h" on page 20747 is in error.]

1. Observations of well formed low temperature ice crystals of around 100 micrometers in size (simple plates and columns) gathered at the ground in the Antarctic do not exhibit trans-prismatic strands (see "Atmospheric Halos" by Walter Tape). Surface distortions which lead to non-optically smooth surfaces (ruining halo production) usually

take the form of layers, steps, and pits or depressions on the scale of 5-10 microns in size that are clearly visible in photographs. The observation of newly frozen drops in the process of forming facets shows similar features at this size scale (Takahashi and Mori, 2006, *Atmos Res.*, **82**, p385-390).

2. Because of the low pressure requirements for operating the SEM, ice crystals were grown at a pressure of around 50 Pa, a factor of 200-800 lower than actual cirrus growth conditions. This extremely low pressure has a marked effect on the vapor diffusivity which is inversely proportional to pressure. Gonda (1976) and Gonda et al. (see *J. Meteorol. Soc. Japan* and *J. Crystal Growth* and references there in) have shown that ice crystal habit is significantly modified by growth at very low pressures like those used by the authors.

3. Crystals were grown in a nearly pure water vapor environment at approximately 500% ice supersaturation which is far in excess of cirrus conditions where ice supersaturations of 25-40% are most common, with values of near 100% recently observed on rare occasions, however, the instrumental accuracy of these measured values is in question. The very high ice supersaturation, together with the extremely large vapor diffusivity in this study, results in exceptionally large crystal growth rates and appears to produce crystal artifacts that are not observed in cirrus ice crystals. The presence of the carrier or background gas (air) at realistic atmospheric pressures is evidently required to accurately reproduce cirrus-like habits and crystal characteristics.

4. The authors comment that the hexagonal plates and columns they observed in their experiments are like those in cirrus clouds. This is incorrect. While simple well formed plates and columns are observed, they are rarely in the majority as evidenced by the scarcity of atmospheric halos. Most cirrus ice crystals are polycrystals with irregular shapes, e.g. multifaceted nugget-like crystals that appear to be quasi-spherical under low image resolution, but are clearly faceted when observed at higher resolution (Bailey and Hallet, *J. Atmos. Sci*, 2009). The concept of spherical ice particles is simply a misconception resulting from poor image resolution. “Warm” cirrus between -40 C and

-60 C can contain significant fractions of columnar crystals, but polycrystalline bullet rosettes are common under these conditions, with irregular, faceted, polycrystals constituting a significant fraction of all crystals observed. These irregular crystals become more prevalent with decreasing temperature (see Lawson et al. *Atmos. Chem. Phys.*, **8**, 1609–1623, 2008).

5. Finally, while the existence of a quasi-liquid layer has apparently been confirmed through ellipsometry techniques at temperatures down to -15 oC (Furukawa et al., Morphology and Growth Unit of Crystals, pp 647-650, Terra Scientific Publishing Co., 1989) and possibly as low as -30 C, there is no evidence that it exists at colder cirrus temperatures, i.e. at low temperatures where even the smallest supercooled droplets cannot exist. I at least am unaware of any studies confirming or inferring the existence of a quasi-liquid layer at temperatures below -40 C, except in theory.

The authors should reformulate the paper as one addressing observations of ice growth under SEM operating conditions, one inferring a possible but suspect relation to in situ ice crystals.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 20739, 2009.