Authors' response to Anonymous Referee #2, RC4
Review received and published: 3 October 2018

For clarity and easy visualization, the referee's comment is copied here in black. The authors' replies are in blue font with an increased indent below each of the referee's statements.

Thanks for providing more information about these experiments. However, authors do not address the concerns that are outlined. I will describe one example here. One of the conclusions of this study (page 5 main paper) is that if no INP was found in a crystal – this crystal was categorized as formed through the process of secondary ice formation. This is based on an observation that this particular crystal (now supercooled droplet) did not freeze until -25C. However, it is possible that this droplet may freeze at colder temperatures than -25C, and if the composition is made up of dissolved organ- ics/inorganics, the droplet may require homogeneous freezing temperatures (< -37C). This possibility is not explored in this study. How to assure that this crystal (or super- cooled droplet) is free of any residue/foreign substance that may trigger nucleation of ice? If the droplet could freeze at < 25C temperatures, then conclusions will change. To verify this possibility an experimental evidence is needed. In response (page 3), it is mentioned that "A possible explanation for the absence of INPs are crystals formed through secondary ice formation processes.", but this is a conclusion which is drawn in this paper based on limited observations, not an explanation. Further, papers from the literature are highlighted saying that low INP concentrations compared to N_ice concentrations are observed previously, but this response does not answer the above question. There are no results regarding the nature of INPs or the freezing spectra of droplets at colder temperatures to understand this concern. My all other questions are somewhat related to this concern. Additional experimental evidence (for example as above) is needed to support the claims made in the paper.

In additional experiments we certainly would find residues or foreign substances in the planar branched crystals we categorise as secondary ice. Such residues could be cloud condensation nuclei in rime droplets, scavenged interstitial aerosol particles, or others. Some of these residues may indeed be capable of triggering ice at temperatures colder than -25 °C. However, initial ice formation at such cold temperatures would not have resulted in the form (habit) of crystals we have analysed. For this reason, we are convinced that they resulted from an ice multiplication process. There is strong evidence supporting this view, which we would include in a revised version of the manuscript:

The temperature range from -20 °C to -70 °C is the so-called "polycrystalline regime" dominated by crystal shapes with a range of different angles between branches or plates extending in three dimensions (Bailey and Hallett, 2009). These crystals will continue to grow when falling into warmer layers of air, as long as these layers are supersaturated with respect to ice. Otherwise, the crystals will sublimate. The growth habit of the falling crystals may change depending on temperature and supersaturation, but it will remain polycrystalline and irregular (c.f. Fig. 6 and 7 in Bailey and Hallett, 2009). Polycrystalline ice particles are highly unlikely to grow into the kind of crystals we have sampled, which had the same angle (60°) between all branches, and branches only extending in a single plane (i.e. dendrites; c.f. Schwarzenboeck et al., 2009).

References

Bailey, M. P., Hallett, J.: A comprehensive habit diagram for atmospheric ice crystals: Confirmation from the laboratory, AIRS II, and other field studies. J. Atmos. Sci., 66, 2888-2899, doi:10.1175/2009JAS2883.1, 2009.

Schwarzenboek,aA., Shcherbakov, V., Lefevre, R., Gayet, J.-F., Pointin, Y., Duroure, C.: Indications for stellar-crystal fragmentation in Arctic clouds. Atmos. Res., 92, 220-228, doi:10.1016/j.atmosres.2008.10.002, 2009.