

We thank the anonymous reviewer for the helpful comments. These comments helped to substantially improve the manuscript. Below we give detailed answers to the individual reviewer comments in blue.

Review of “Cloud chamber experiments on the origin of ice crystal complexity in cirrus clouds” by Schnaiter et al.

Recommendation: Accept after minor revision

This paper reports on the origin of ice crystal complexity and its influence on the angular light scattering properties of cirrus clouds based on cloud simulation experiments in the AIDA cloud chamber. Ice particles were grown by both homogeneous and heterogeneous nucleation, and subsequently grown and sublimation at super and sub-saturated conditions. Ice crystal complexity was subsequently deduced from light scattering patterns measured by the SID-3. The principle finding was that ice crystal complexity is dependent on the available water vapor, and that this complexity dominates the microphysics. As with observations from natural clouds, the measured scattering phase functions were featureless resulting in low asymmetry factors that differ from those of several idealized crystals that are used to make up common scattering libraries. I think the paper is appropriate for publication in ACP because it is the first effort that shows ice crystal complexity is correlated to the available water vapor, and shows that some of the featureless phase functions observed in nature can be replicated in a laboratory environment. Nevertheless, I think there are some aspects of the presentation that could be improved before the paper is accepted for publication. Many of these aspects have already been identified by the other reviewers of the paper, so I will restrict my comments to a few other aspects of the presentation and emphasize a couple of the points previously made.

The major comment I would make is that crystal complexity needs to be better defined or better described. There are many aspects of ice crystals that affect their complexity and how they affect radiation from their three-dimensional shapes and their complexity, to the small-scale surface roughness to variations in aspect ratios. I think crystal complexity goes much beyond surface roughness as is described in the introduction. I don't think this paper ever formally describes what exactly is meant by crystal complexity or gives any perspective about the importance of complexity in affecting radiative properties compared to other microphysical properties (e.g., aspect ratio, size, shape, etc.). I would recommend that such material be added to the introduction.

We agree with the reviewer that the definition of crystal complexity was not given in a nuanced way. Therefore, we have changed our complexity definition to “all surface distortions on a single ice particle (surface roughness on a variety of scales, polycrystallinity, and hollowness)”, and named it “small-scale complexity”. In this way, we think there is a clear differentiation to large-scale complexity that is induced by crystal aggregation and that can be easily observed by high resolution imaging (e.g. Schmitt and Heymsfield, 2014).

Related to the above point, details about the modeling work and the parameter k_e are currently found in Appendix B. As this paper is about crystal complexity and k_e is found to be the most robust feature

parameter to characterize crystal complexity with the SID-3, I think this modeling work should not be relegated to an Appendix but rather should appear in the main body of the paper.

We were stimulated by the work of Lu et al. (2006) who concluded that k_e is the most robust measure of surface roughness. We applied this measure to the SID-3 scattering patterns in order to characterize ice crystal complexity. A clear description of k_e , what it means in terms of a physical roughness, and how it is determined from the SID-3 patterns is now presented in Section 2.1.2 of the revised manuscript. The robustness of this measure in case of single particle scattering patterns was tested with analogue ice particles as described in Appendix A. We added the result of this test to Section 2.1.2 of the revised manuscript.

A first rough correlation between the complexity parameter k_e and the geometrical particle complexity was found in the modeling study for deformed Gaussian spheres shown in Appendix B. We are aware of the fact that ice particles are in general faceted (despite frozen droplets for which the Gaussian sphere model might be more realistic (Nousiainen et al., 2011)). Therefore, it is our intention to test the SID-3 complexity analysis with more realistic optical models for hexagonal cirrus ice particles in the future. That is why we think that the presentation of this modeling study should stay in the Appendix rather than being moved to the main body of the manuscript.

Nousiainen, T., Lindqvist, H., McFarquhar, G. M., and Um, J., 2011: Small Irregular Ice Crystals in Tropical Cirrus. *Journal of the Atmospheric Sciences*, 68(11), 2614–2627.
<http://doi.org/10.1175/2011JAS3733.1>

In terms of experimental procedure, the authors claim that a sublimation period was applied in order to remove the ice particle surface characteristics from the initial growth. How confident are they that they are removing all of these surface characteristics? It would seem that apart from a derived measurement of the scattering function, it would be very difficult to know how these surface characteristics are changing. In addition to the effects investigated by the authors, would any small scale imperfections in the ice nuclei have impacted the surface roughness, complexity, and scattering patterns as well?

As stated in step 4 of the procedure list given in Section 2.2.2, sublimating conditions are applied for a period long enough to see a significant change in the SID-3 scattering patterns. As we know from the characterization of the SID-3 complexity method (e.g. Fig. B1) the SID-3 scattering patterns are very sensitive to smallest changes of the surface properties. What we typically observe during the sublimation periods is a gradual change of the acquired scattering patterns to oval patterns as shown in the upper panel of Fig. 3. The appearance of oval patterns is not only restricted to pristine hexagonal columns as depicted in Fig. 3, but is also observed in case of complex speckled patterns. As a subsample of the acquired SID-3 scattering patterns are displayed online by the acquisition software, a visual monitoring of these patterns by the experimentalist during the sublimation period gives detailed information on how the particle surface properties are changing with time and if the sublimation period has applied long enough. This is now described more detailed in the revised version of the manuscript.

As mentioned in the Conclusions, we do not see a significant difference in the ice crystal complexity for the different heterogeneous ice nuclei. However, there are indications for a memory effect in the case of homogeneous nucleation in aqueous sulphuric acid particles. This first observation will be further investigated in upcoming chamber experiments.

Page 30513, line 16. Although the net radiative effect of a single cirrus cloud can be altered by this much, the global effect as a whole is much less. I think this point should be made more clear so as to avoid over stating the impact of complexity.

We extended the corresponding sentence by “..., though the global effect by cirrus clouds is much less.”

I would recommend that a photo of the experimental setup be added to the manuscript. That gives more of a visual description of how the experiment was setup. Alternatively, a schematic of the experimental setup would suffice.

A schematic of the experimental setup can be found in Schnaiter et al. (2012). As there are no significant changes to this configuration, there is no need to show the same schematic again. However we added a reference to the Schnaiter et al. (2012) schematic to Section 2.2.1.

Page 30520, line 19: Is there any bias to the sample by restricting to images with a narrow mean brightness range between 10 and 25? Were any sensitivity tests done to determine the effect of broadening?

Broadening the brightness range has a small influence on the k_e analysis in a way that e.g. the rough particle fractions given in Tab. 1 would increase by a few absolute percent when lowering the lower limit from 10 to 5. The upper limit in brightness range has less an influence.

Page 30522, line 21: The speed of 10 m/s is substantially smaller than that which would be used in an aircraft flight campaign where many prior scattering phase functions were measured. Is there any impact of the flow velocity on your results?

We do not expect an influence due to the lower particle speed as the exposure time for the SID-3 image capture is not controlled by the residing time of the particle in the laser beam but is set to a fixed value of 2 μ s.

Page 30523, last 2 paragraphs: Were any other cloud probes also used to make measurements during the cloud expansion runs? This might provide an interesting data set for comparison against the scattering functions.

In part of the experiments we also operated other cloud probes. Comparisons of the measurement results from those instruments with the light scattering measurements will be the subject of forthcoming papers.

Page 30524, line 5: Can you quote the uncertainty on water vapor measurements here since this is a critical parameter for interpreting the results?

The water vapor measurement in terms of the partial vapor pressure has an accuracy of better than $\pm 3\%$ (Fahey et al., 2014). This value is given in the revised manuscript.

Page 30524, line 19: how much uncertainty was there in the saturation with respect to ice, and how repeatable were the measurements?

The accuracy of the saturation ratio with respect to ice is dominated by the accuracy of the water vapor measurement and, therefore, is in the range of ± 0.03 (Wagner et al., 2010). This value is given in the revised manuscript.

Page 30527, line 2: Define all acronyms.

Acronyms are defined in the revised manuscript.

Apart from these minor points, I think this paper is acceptable for publication in ACP.