

This paper provides new insights into the relationship between atmospheric state variables and ice crystal complexity through the novel and original use of cloud chamber experiments. Previous cirrus in situ-based work has failed to find such relationships because real atmospheric ice particles undergo different cycles of subsaturation and supersaturation, resulting in weak evidence. This laboratory-based paper reports results based on well controlled experiments of single cycles of subsaturation and supersaturation, with respect to ice, measured over some interval of time. As a result of this more systematic approach, they find correlations between ice crystal complexity and atmospheric state variables. At the same time, as these controlled experiments SID-3 two-dimensional light scattering patterns are obtained, and these were used to estimate surface roughness or ice crystal complexity, which could then be related to supersaturation w.r.t ice. They report a positive correlation between ice crystal complexity and atmospheric state variables. They also make good use of an electromagnetic method by applying it to a distortion of the sphere to derive the speckled 2D patterns measured by SID-3. By increasing the model distortion value they show that this too is related to their proxy for surface roughness, and hence atmospheric state variables. Moreover, a previously fitted phase function is applied to their laboratory results and from that they deduce an asymmetry parameter value of 0.78. A useful light scattering modelling result to come out of this paper is the experimental confirmation that once the ice particle is sufficiently randomised, the resulting measured light scattering pattern is invariant with respect to shape. Finally, they generalise their laboratory-based findings to the real atmosphere. This paper is an important step along the way to understand why atmospheric ice particles are observed to be generally complex. This paper should be published, but only after the following points have been considered and discussed.

1. There are numerous examples of typos, incomplete sentences, and incorrect words. Please could the authors more thoroughly proof read their paper before resubmitting a revised version?
2. The authors define ice crystal complexity as “...any kind of crystal distortions (surface roughness, polycrystals, aggregates, (stepped) hollowness).” and yet their measure of complexity is best related to surface roughness according to Lu et al. (2006). That paper does not state anything about the dependence of  $k_e$  on any of the other variables listed in their definition of ice crystal complexity. Therefore, are the authors saying that their SID-3 laboratory measurements are more related to surface roughness than the other variables listed? Given the size range listed, and their figure 5 caption, which shows no aggregates or polycrystals and states “...surface properties are masked by the Formvar replication method...”. Given the above, it seems to this reader that their results are indeed more related to surface roughness and so for this paper their definition of ice crystal complexity is redundant? They have yet to show the dependence of the SID-3 light scattering patterns on all the other variables inclusive of surface roughness. This is an important point, as it is necessary to show whether surface roughness alone is sufficient to replicate the SID-3 measurements, even if the particles might also be hollow, polycrystalline, rosettes, plates or aggregate combinations of some or all of these shapes.
3. The discussion in the main text is related to  $k_e$  and not the parameter referred to as the “image texture feature energy” in the caption of Figure 1. What

exactly is the latter? Since they use the former, perhaps the latter could be removed? If it is important how else is it used? Its quantitative use is not at all clear?

4. In the introduction, page 6, line 5. The authors only discuss surface roughness with regard to the uncertainties in cirrus radiative effects in a climate model. There are of course a number of other important cirrus properties that contribute to this uncertainty apart from surface roughness. Surface roughness is important, but there are a number of other properties that must also be considered, and the uncertainties associated with these may have the greater impact on cirrus radiative effects. These are the amplitude and distribution of the small ice mode, the shape of the PSD, the distribution of shapes across the PSD, and the shape distribution as a function of distance from the cloud-top. The authors should cite the following papers that discuss all the above (there are of course others, but these are representative) and these are listed as follows: (1) Mitchell, D. L., P. Rasch, D. Ivanova, G. McFarquhar, and T. Nousiainen (2008), Impact of small ice crystal assumptions on ice sedimentation rates in cirrus clouds and GCM simulations, *Geophys. Res. Lett.*, 35, L09806, doi:10.1029/2008GL033552, (2) Anthony J. Baran, Peter Hill, Kalli Furtado, Paul Field, and James Manners, 2014: A Coupled Cloud Physics–Radiation Parameterization of the Bulk Optical Properties of Cirrus and Its Impact on the Met Office Unified Model Global Atmosphere 5.0 Configuration. *J. Climate*, 27, 7725–7752. doi: <http://dx.doi.org/10.1175/JCLI-D-13-00700.1>, and (3) Yang, H., Dobbie, S., Herbert, R., Connolly, P., Gallagher, M., Ghosh, S., Al-Jumur, S. M. R. K. and Clayton, J. (2012), The effect of observed vertical structure, habits, and size distributions on the solar radiative properties and cloud evolution of cirrus clouds. *Q.J.R. Meteorol. Soc.*, 138: 1221–1232. doi:10.1002/qj.973.
5. In the introduction, page 4, 3<sup>rd</sup> paragraph, line 4. Once again, in my opinion, there needs to be more of a balance in the choice of citations, as the citations chosen appeared after others had already shown some of their results. The discussion is essentially about the consistency of models using observations from across the spectrum and the evidence so far tends to show that randomised models are better at simulating simultaneous multi-spectral, multi-angle observations. Other citations that ought to be included are listed as follows: (1) Baran, A. J. and Francis, P. N. (2004), On the radiative properties of cirrus cloud at solar and thermal wavelengths: A test of model consistency using high-resolution airborne radiance measurements. *Q.J.R. Meteorol. Soc.*, 130: 763–778. doi:10.1256/qj.03.151, (2) Baran, A. J., Havemann, S., Francis, P. N., and Watts, P. D. A consistent set of single-scattering properties for cirrus cloud: tests using radiance measurements from a dual-viewing multi-wavelength satellite-based instrument. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 79-80, 549-567, 2003, and (3) Baran, A. J., Cotton, R., Furtado, K., Havemann, S., Labonnote, L.-C., Marengo, F., Smith, A. and Thelen, J.-C. (2014), A self-consistent scattering model for cirrus. II: The high and low frequencies. *Q.J.R. Meteorol. Soc.*, 140: 1039–1057. doi:10.1002/qj.2193.

6. In the introduction, page 4, line 12. Field et al. (2003) used mid-latitude SID-2 measurements to test the angular scattering properties of models, so this paper should also be cited. In Field et al. (2003), no evidence for halos was found. Field, P. R., A. J. Baran, P. H. Kaye, E. Hirst, and R. Greenaway (2003), A test of cirrus ice crystal scattering phase functions, *Geophys. Res. Lett.*, 30, 1752, doi:10.1029/2003GL017482, 14.
7. As stated in point 4 above, determining the small ice mode is important and the authors of this paper are aware of this problem. Unfortunately, in this paper, there are no comparisons of PSDs measured by the differing instruments. Please show examples of such comparisons. The speckle pattern reported in this paper could also be used to determine ice crystal size as shown by Ulanowski et al. (2012) [JQSRT 113, 2457-2464]. How does this method compare to independent measurements of small ice?
8. An example of one of the limitations of this study scaled to the real atmosphere is the reported infrequent occurrence of hollow ice crystals. However, in situ observations by Schmitt and Heymsfield (2007) [Schmitt, C.G., and A.J. Heymsfield, 2007: On the occurrence of hollow bullet rosette- and column-shaped ice crystals in midlatitude cirrus. *Journal of the Atmospheric Sciences*, 64, 4514-4519, DOI: 10.1175/2007JAS2317.1] show that hollow ice crystals can frequently occur at cirrus forming temperatures. Moreover, the cloud chamber study of Smith et al. (2015) [Smith, H R, Connolly, P J, Baran, A J, Hesse, E, Smedley, A R D & Webb, A R 2015, Cloud chamber laboratory investigations into scattering properties of hollow ice particles, *Journal of Quantitative Spectroscopy and Radiative Transfer*, vol 157, pp. 106-118, ., 10.1016/j.jqsrt.2015.02.015] report that stepped hollow columns can reduce the asymmetry parameter, relative to other forms of cavity that are usually assumed, and these occurred frequently in their experiments at temperatures down to -30°C. Therefore, the geometric form of cavity assumed will affect the asymmetry parameter. The current studies are limited to a narrow size range and ppmv values, and so cannot be expected to cover the size range measured in the studies reported above. The authors should discuss in more detail the limitations of scaling their laboratory results to the real atmosphere.
9. Section 3, page 18, line 23. The authors report a relationship between ice crystal complexity and supersaturation. Recently reported satellite-based observations by Baran et al. (2015) [Baran, A. J., Furtado, K., Labonnote, L.-C., Havemann, S., Thelen, J.-C., and Marengo, F.: On the relationship between the scattering phase function of cirrus and the atmospheric state, *Atmos. Chem. Phys.*, 15, 1105-1127, doi:10.5194/acp-15-1105-2015, 2015] tend to support this view. However, such a strong link could not be found statistically, due to there being too few cases. Nevertheless, in Figure 11 of that paper, it can be seen that pixels related to phase functions exhibiting ice bow features were more associated with low NWP model supersaturation values. Whilst pixels associated with phase functions exhibiting no features at backscattering angles were mostly associated with supersaturation values  $\gg 1$ . Note also, the range in asymmetry parameter values reported in that paper was between 0.82 and 0.79. A 5% difference in the asymmetry parameter is climatically

important. This is why we need to understand the relationship between ice crystal complexity and the atmospheric state.

10. The question arises as to how such a relationship can be incorporated into climate models? This paper provides the first steps towards this eventual aim through Figures 4 and 6. However, there does need to be a comment about Figure 6. From this figure, it would seem that for mixing ratios  $> \sim 5$  ppmv complex particles can be assumed or roughened particles, and from Figure B2 this occurs at distortion values  $> \sim 0.4$ , and Fig. B1 suggest that surface amplitude irregularities need to be significant. However, this value of 5 ppmv is very small within a climate model. Are the authors sure that this figure is correct? Or rather are the numerical values along the x-axis correct? The authors use ppmv, whereas in atmospheric models mixing ratio is usually in units of kg per kg and 5 ppmv translates to about  $\sim 10^{-6}$  kg per kg? In some atmospheric models, this value is taken as the threshold value for the existence of cloud. Therefore, the authors imply that in atmospheric models, the ice particles should always be rough as non-roughened ice particles cannot exist in such models. At least according to their threshold value or is there an error somewhere? This could be yet another limitation of this experiment to the real atmosphere? Please comment. Moreover, why not on the x-axis just plot the results in kg per kg? As this unit is more directly related to atmospheric models whereas ppmv is more related to chemistry.
11. The experiments of substuration and supersaturation are over a single cycle. What would happen to the ice particle complexity if several cycles were measured? In the real atmosphere, cirrus ice particles undergo a number of cycles, and this might completely change their level of complexity such as the formation of polycrystals, and these in turn, will change their scattering properties and g-values. Please comment and discuss.
12. Figure B1. Please can the authors, for each assumed distortion value, provide the corresponding model area ratio and particle effective density? How well do these values compare to more recent observation of small ice area ratios and effective densities? More recent observations of these parameters can be found, respectively, in the following list of papers: (1) Greg M. McFarquhar, Junshik Um, and Robert Jackson, 2013: Small Cloud Particle Shapes in Mixed-Phase Clouds. *J. Appl. Meteor. Climatol.*, 52, 1277–1293 doi: <http://dx.doi.org/10.1175/JAMC-D-12-0114.1> and (2) Cotton, R. J., Field, P. R., Ulanowski, Z., Kaye, P. H., Hirst, E., Greenaway, R. S., Crawford, I., Crosier, J. and Dorsey, J. (2013), The effective density of small ice particles obtained from in situ aircraft observations of mid-latitude cirrus. *Q.J.R. Meteorol. Soc.*, 139: 1923–1934. doi:10.1002/qj.2058. Just because the assumed idealised model can be made to produce a speckled pattern does not mean that the model is consistent with the most recent observations of microphysics in terms of area ratio and mass. These parameters are also important to cirrus radiative transfer. Another idealised model, based on varying the Chebyshev model along the directions of theta and phi, which could also be considered by the authors is described here <http://www.ncbi.nlm.nih.gov/pubmed/21716343>. Does this model also produce speckled patterns? It would be interesting to see.

13. A comment on Figure 7. Extrapolating the fit to this figure to deduce the asymmetry parameter is not convincing. Moreover, there are no observations at scattering angles  $\ll \sim 20^\circ$  and at scattering angles  $> \sim 158^\circ$ . It is quite possible to arrive at an alternative extrapolation to the one provided (by simple examination by eye of the figure), and this would give a completely different value for the asymmetry parameter. Their extrapolation is not supported by measurements at lower or higher scattering angles as all scattering angles are required to deduce the asymmetry parameter. Unfortunately, from this figure, it is not possible to come to a definite conclusion about a value for the asymmetry parameter. Let alone extrapolate it to the real atmosphere as the figure is related to their experiments having only undergone single cycles. Moreover, the phase function structure predicted by the assumed model at scattering angles greater than  $160^\circ$  is not supported by the most recent multi-angle global observations of cirrus. See, for example, the paper at this link <http://www.atmos-chem-phys-discuss.net/15/31665/2015/acpd-15-31665-2015.html>. In particular, note that Figure 2a in the above paper shows that the PDF of sampled direction peaks at scattering angles at around  $160^\circ$ , and it is still significant at  $170^\circ$ . Figure 7, in the same paper, shows that models exhibiting significant phase function structure in the backscattering direction near to  $180^\circ$  do not satisfy the observations. The model of the phase function shown in Figure 7 cannot be extrapolated to the real atmosphere.

However, having said that, it is still important that in-situ probes not only measure the scattered intensity at forward scattering angles but also at backscattering angles near to  $180^\circ$ . The authors may wish to elaborate on this point in their paper. This point is also made by Baran et al. (2012). If measurements were available at near  $180^\circ$  backscattering angles, then the model shown in Figure 7 might have been rejected.

14. Why do the authors not show a time series of the SIMONE linear depolarization ratio covering the subsaturated and supersaturated cycles? This measurement seems to be briefly mentioned in their paper. Why not use this SIMONE measurement to test the model at backscattering angles near to  $180^\circ$  shown in Figure 7? Please comment and show.

Given the above caveats about extrapolating the experimental results to the real atmosphere the discussion in section 4 needs to be substantially revised. Some of the references used in section 4 probably can no longer be used as support for their extrapolations simply because the cirrus microphysics references are highly likely to have suffered from the shattering of ice crystals on the inlets of the microphysical probes, and this problem is not discussed at all in the section and how it might have affected the results.

