

**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.**

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?

**We will thoroughly proof read the revised paper before submission.**

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.

**In the presented experiments, we have only single particles present, so, particle aggregation can be excluded from the list of complexity types that influence  $k_e$ . Polycrystallinity and hollowness, on the other hand, are clearly present in our ice particle populations (Fig. 5). As  $k_e$  is a measure of the degree of spatial randomization of the scattered light intensity, it is currently not possible to discriminate these complexity types from surface roughness without any further information. For example, the speckle patterns produced by fine-roughness mineral dust grains and highly complex ice analogue crystals are similar, as previously shown (Ulanowski et al., 2014). However, in the case of hollow columns there are indications of a specific triangular feature around the 22° halo**

spots of the scattering patterns (visible on the second pattern of row (b) in Fig. 1), which might be useful to discriminate this complexity type in future.

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).

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?

We agree with the reviewer that the “energy feature” does not add any further information and actually isn’t used for data interpretation in the manuscript. So, we omitted the energy feature from the image and caption of Figure 1.

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>

(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.

We agree with the reviewer and added the following sentence to page 5, 1<sup>st</sup> paragraph: “As already mentioned, the uncertainties in assessing the radiative effect by cirrus clouds are due to the uncertainties of many macroscopic as well as microscopic cloud properties like IWC, the vertical structure of the ice particle size and shape distributions, the amplitude and distribution of the small ice mode, etc. (Mitchell et al, 2008; Yang et al., 2012; Baran et al., 2014).”. However, we would like to emphasize that although the prediction of cloud formation and evolution is important, the optical properties of the constituent ice particles eventually define the radiative effect of the cloud once it has been formed.

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

(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.

We agree with the reviewer and have added the suggested references accordingly.

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.

We added the following sentences to page 4: “Field et al. (2003) analyzed SID-2 single particle scattering patterns in order to draw conclusions on the scattering phase function of cirrus ice crystals. They found no evidence for 22° halos in their data set of mid-latitude cirrus.”

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?

We agree that the small ice particle mode is important for the radiative properties of cirrus clouds and the measurement of this microphysical property is highly uncertain due to the different methods used by in-situ cloud probes. In the AIDA chamber, due to the experimental conditions, we know that (i) we only have ice particles smaller than 50  $\mu\text{m}$  and (ii) this small ice particle mode is not biased by particle shattering. Therefore, such experiments are extremely useful in comparing in-situ cloud probes although this was not the primary focus of this work. To give the reader an impression of the potential we have in this respect, we added the result of the PHIPS-HALO image analysis to panel (c) of Fig. 2. In doing so, this panel now shows a comparison of two different particle sizing methods; the laser scattering method used in PPD-2K and SID-3 and the imaging method used in PHIPS-HALO. Although the evolution of the size distribution agrees very well, there is a systematic size difference between the two instruments.

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  $-30^{\circ}\text{C}$ . 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.

It is possible that the reviewer might have misinterpreted our results. The term “occasional” is related to the observation of stepped hollowness that we classified as surface roughness type of complexity. The fact that stepped hollowness was infrequently observed in our experiments is rather due to the limitations in the used imaging methods - a high level of magnification is required to resolve stepped hollowness - then being a property of the particle ensemble. In fact, we observed hollowness quite frequently in the imaging data (replica and PHIPS) especially in the  $-50^{\circ}\text{C}$  runs and for  $S_{\text{ice}}$  above 1.15 and it is likely that at least a part of this hollowness is stepped. Therefore, we think that our results are in accordance with the studies mentioned by the reviewer and are of relevance for the real atmosphere.

We will discuss our hollowness observations more clearly in the revised paper to avoid further misinterpretations.

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.

We agree with the reviewer that the mentioned satellite-based study supports one of our main findings from the laboratory, namely a correlation between ice crystal complexity and supersaturation. Therefore, we added a paragraph to Section 4, in which we discuss our laboratory results in the context of the study by Baran et al. (2015).

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.

Here, we have to emphasize that in Fig. 6 we have plotted the median roughness parameter over the available condensable water vapor mixing ratio. The latter is not the absolute water vapor mixing ratio as it is usually calculated by numerical weather prediction models, but the mixing ratio of those molecules that are in excess with respect to the molecular number concentration at ice saturated conditions, i.e. the difference between the absolute mixing ratio and the mixing ratio at ice saturated conditions for the actual pressure and temperature in the chamber. Therefore, this value is inherently zero at ice saturated conditions. We introduced this quantity here because we wanted to compare the complexity results of the two sets of experiments performed at initial temperatures of  $-40^{\circ}\text{C}$  and  $-50^{\circ}\text{C}$  in Figure 6 without the superimposed ice saturation vs. temperature dependence that is inherent in Figure 4. We believe that this is explained in a clear and sufficient way in Section 3.1 of the manuscript.

The reviewer is correct, when pointing out that the study presented here is the first step to understand the formation of crystal complexity of atmospheric ice particles. However, further work is still needed to implement these results in climate models.

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.

As already mentioned in the description of step 5 of the growth and sublimation cycles (Sect. 2.2.2), several regrowth cycles can be performed within the same cloud provided that a significant ice particle number concentration is left in the chamber from the previous cycle. Actually, nine of the data points shown in Figs. 4 and 6 are from higher order regrowth cycles. Typically, these higher order cycles were performed at higher supersaturations than the previous cycles, just because the ice crystal concentration is decreasing with time and therefore higher supersaturation levels can be reached at later times. These data points nicely fit to the general trends given in Figs. 4 and 6, i.e. there are no indications that a possible formation of polycrystals with time has a significant influence to the results. However, there is one cycle where the second regrowth was performed at lower  $S_{\text{ice}}$  (1.04 vs. 1.18 as in the first cycle). This cycle indeed results in a slightly higher complexity parameter ( $k_e=4.49$ ) of the ice crystals compared to first order regrowth experiments at similar  $S_{\text{ice}}$  ( $k_e\approx 4.3$ ). In conclusion, the formation of polycrystals with time might result in a slight increase of the crystal complexity within the ice cloud, but this does not change the general results and conclusions of the study.

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>

(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.

We have calculated the area ratio  $\alpha$  and the effective density  $\rho_e$  of the model particles shown in Fig. B1. These values are given in the revised version of Fig. B1 (now Fig. ??? in the revised manuscript). These values are well in agreement with the more recent observations of  $0.6 < \alpha < 0.8$  for small ( $< 50 \mu\text{m}$ ) ice particles in Arctic ice clouds (McFarquhar et al., 2013) and  $\rho_e=700 \text{ kgm}^{-3}$  for small ice particles in mid-latitude cirrus clouds (Cotton et al., 2013). Therefore, our idealized optical model is consistent with the most recent microphysical observations and it is justified to use this model to get a first rough correlation between the complexity parameter  $k_e$  deduced from SID-3 and the geometrical particle complexity. 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. Although it would be interesting, testing other optical models (like the suggested Chebyshev model) is beyond the scope of this paper.

Here, we would like to emphasize an important point in the interpretation of microphysical observations. As has been shown in a conference contribution by Ulanowski et al. (2004) <http://vuh-la-rispr.herts.ac.uk/portal/services/downloadRegister/426101/101433.pdf> (Figs. 5 and 6), the microphysical properties of small near-spherical ice crystals determined from CPI images (as used in the study by McFarquhar et al., 2013) do not necessarily give information on the small-scale crystal complexity. This means that also in recent microphysical observations, part of the crystal complexity is hidden due to resolution limitations of imaging cloud probes. It has been recently shown by Ulanowski et al. (2014) and Järvinen et al. (2016) that this hidden small-scale crystal complexity can dominate the light scattering properties of small, near-spherical ice particles.

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.

We agree with the reviewer that the determination of the asymmetry parameter from a comparison of the nephelometer measurements with a published model scattering function is not justified. Although this might be possible to some extent - either by extrapolating the missing forward scattered intensity based on the method presented by Gerber et al. (2000) or by using the iterative inversion method developed by Oshchepkov et al. (2000) and Jourdan et al. (2003) - the main conclusions from Fig. 7 as listed and discussed in Section 3.3 are still valid. Additionally, as already mentioned in the answer to the reviewer comment 11, part of the presented results are from multi-cycle growth experiments. We therefore believe that our results are of high relevance for the real atmosphere. However, we see that the reader might be misguided by giving the asymmetry parameter of the model scattering function in the discussion of Fig. 7 in Section 3.3, and we will revise this section as well as the Abstract and the Conclusions accordingly.

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.

It is not yet possible to use the SIMONE backscattering results to test the scattering model shown in Fig. 7 because this requires an absolute measurement of the scattered intensity which is not yet implemented. However, there are some interesting results of the SIMONE linear depolarization measurements with respect to ice particle complexity, which will be the subject of a forthcoming paper.

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.

We agree and will revise section 4 accordingly.