Authors' Response to Anonymous Referee #1 Comments

The authors would like to thank the anonymous Referee for her/his comments that have helped us to improve this manuscript. Below, the major and minor comments are addressed by detailed point-by-point replies. Referee's comments are in blue and authors' replies in black.

General comments

The paper presents a compilation of light scattering measurements obtained from a large number of aircraft campaigns, distributed globally, and they relate these measurements to ice crystal submicron complexity. This enables the authors to obtain estimates for the asymmetry parameter, a parameter of importance in NWP and climate modeling. They find from their analyses that the asymmetry parameter determination of 0.75 can be related to their complexity findings. This appears invariant with location and ice/cirrus formation, and the resulting scattering pattern results from the observed ice crystal complexity. As a consequence, this complexity expressed through the asymmetry parameter induces a not insubstantial-averaged further cooling effect not currently accounted for in climate models.

This is a largely well written paper, which links experimental results with theory and relates these measurements to ice crystal complexity and follows the theory through to an application in climate models. The paper provides nice results which deserve to be published, but the claim needs to be proven more rigorously with uncertainties attached to their estimates.
We thank the Referee for this positive general comment. In her/his comments the Referee has raised concerns on the rigour of the presented conclusions, especially on the uncertainties in the asymmetry factor. We acknowledge that the limitations of our measurements were not adequately discussed and have modified the discussion and conclusions to be more sensitive to these limitations. Below are listed the detailed replies to the Referee's major and minor comments.

Major comments

1. The claim of the authors is that their measured PN angular scattering patterns are sufficient to determine the asymmetry parameter through some theoretical phase function that appears to fit through the data. This is not convincingly shown to be the case and appear to be eye fits at one single wavelength. There is no discussion in the text as to how the best fit to the measurements was statistically determined? Moreover, there are a number of extrapolations that could be used owing to the spread throughout the data, what uncertainty does this spread produce in the estimated asymmetry parameter values? There should be an uncertainty attached to their estimate of 0.75±? Once these uncertainties have been derived for the asymmetry parameter, the uncertainty in the SWCRE should be consequently determined.
Estimating asymmetry factor from measurements that are covering only part of the angular region is challenging, especially since majority of the scattered intensity is found in the forward direction that is not covered by the measurements. We think that the best approach to estimate the asymmetry factor from the measurements is to use a physical model with a known asymmetry factor to fit the measurements in the known angular range, as done in this manuscript.

The same difficulty applies to estimating the uncertainty of the asymmetry factor if only part of the angular range is covered. For example, if we calculate the partial asymmetry factor for the angular range of 18 to 170° using the column aggregate model, we get an partial asymmetry factor of 0.14. This is approximately 20% of the total asymmetry factor of 0.75, since the forward peak contributes the majority part of the asymmetry factor. Even if we can estimate the uncertainty between the fit and the measurements in the restricted angular range, this uncertainty estimation contributes only 20% to the total uncertainty. Therefore, the measurements alone are not enough to estimate the uncertainty in the asymmetry factor, although it can be argued that the uncertainty of the asymmetry factor is the highest in the measurement region compared to the forward region, where the scattering intensity is mainly determined by the particle size and less of shape.

Owing to this discussion, we agree with the Referee that the discussions in the Sect. 3.4 and in the abstract and some of the conclusions are not well justified. Better than *retrieving* the asymmetry factor from the measurements it is more justifiable to *compare* the different optical models to the measurements. Therefore, we have reformulated the Sect. 3.4 and the section title as “Comparison of the measured angular scattering functions to a light scattering database”. We have also omitted the sentence “*Using the severely roughened hexagonal aggregate model asymmetry factors of 0.750 and 0.754 at 532 nm and 804 nm, respectively, were retrieved*” and instead write in the Sect. 4: “*the severely roughened hexagonal aggregate model has relatively low asymmetry factors of 0.750 and 0.754 for 532 nm and 804 nm, respectively*”. The abstract we have modified so that instead of writing: “*as a consequence, a low asymmetry factor of 0.75 is observed*”, we write “*as a consequence, a similar flat and featureless angular scattering function is observed. A comparison between the measurements and a database of optical particle properties showed that severely roughened hexagonal aggregates optimally represents the measurements in the observed angular range*”.

We also make a stronger case in the Sect. 3.4 that the severely roughened hexagonal aggregate model best represents our measurement. We have added a new figure that compares the different models with the measurements at 804 nm (please see answer to comment 2) and justify the fit by calculating the root mean square errors (RMSE) between the model and the mean of the measurements. For both wavelengths the column aggregate model has the lowest RMSEs of 0.0017 and 0.0014 (for 532 and 804 nm, respectively) compared to the other models (RMSE between 0.0022 and 0.0111 for 532 nm, and 0.0037 and 0.0208 for 804 nm). The discussion of the RMSE analysis was added to the Sect. 3.4.

The asymmetry factors around 0.75 are, therefore, not *retrieved* from the measurements but represent the asymmetry factor of the severely roughened hexagonal aggregate model. This asymmetry factor is fixed for a given size distribution. Later, we use the severely roughened hexagonal aggregate model for deriving the parameterization of SW asymmetry factors for the ECHAM-HAM model. We believe this approach is still justified based on the microphysical observations. However, we agree that the claim in the last sentence of the first conclusions paragraph is not well justified and this conclusion is rewritten in the revised version (please see the reply to comment 3).

2. The other wavelength of 0.804 um is only once shown, the same as Figure 5 should be shown but for 0.804 um using all models. Moreover, the eight-column aggregate shown at 0.804 um, is only just within the measured uncertainties at side scattering angles. This could be owing to the aspect ratio of the monomer columns not being sufficiently large and spaced out more than the compact model they show. The aspect ratio is also an important determinant of the asymmetry parameter as shown by Fu (2007), among others. It would be interesting to plot the approach of Fu (2007), to see if that treatment provides similar low values to those being estimated from the data.

We added a new figure (Fig. 6) showing a comparison between the PN measurements and different models results calculated for 804 nm. This comparison shows that from all of the different habit models, the severely roughened column aggregate model has the best overall agreement with the measurements at 804 nm. Other models underestimate the backscattering intensity from 120° onwards. A discussion of this comparison was added in Section 3.4.

The Referee also suggests to modify the aspect ratio of the severely roughened column aggregate model in order to find a better fit to the measurements at the 804 nm wavelength, as done in the work of Fu (2007). Fu (2007) has showed that modifying the aspect ratio will influence the asymmetry factor of single hexagonal columns. We agree that modifying the aspect ratio of the severely roughened column aggregate model to create a better fit at 804 nm would be an interesting investigation. However, modifying the existing optical particle model or introducing new optical models is not the scope of this paper for two reasons. First, the focus of this paper is to present globally distributed observations of ice crystal mesoscopic complexity and relate them to the angular light scattering measurements. The modelling efforts in this paper are used as a tool to understand the implications of the measurement results. We hope that the measurements will inspire to development of new optical particle models in the future. Secondly, we do not have enough spectral information on the angular scattering functions that we can justify using different optical models for different wavelength bands. We think that the best approach here is to use one optical model to calculate the asymmetry factors for all the SW bands. The use of the severely roughened column aggregate model is justified since it provides the best overall fit on both wavelengths.

3. The paper concludes that it is appropriate to apply the eight-column aggregate in climate and weather models. This is a rather significant claim as the model has only been tested at one single wavelength, at 0.805 um, it does not appear to possess the correct absorption properties at side scattering angles for the possible reasons stated above. It is unclear as to how this model would fit observations at other wavelengths of importance, such as in the terrestrial window region, far infrared, and at more absorbing solar wavelengths, such as at 1.6 and 2.2 um. These wavelengths are also of importance in weather and climate modelling. The authors present no evidence to support their general claim.

We agree that the spectral consistency of optical particle models is one of the biggest challenges in current climate models and in remote sensing retrievals. Modelling the spectral dependency of the asymmetry factor is difficult, since atmospheric measurements are available in only few wavelengths. Additional challenge is posed through the fact that each of the operated polar nephelometer work at a single wavelength, and therefore, combining the polar nephelometric measurements to gain spectral information will inevitably contain uncertainties merging from different measurement setups. For example, in our case we cannot completely distinguish, which proportion of the difference we see between the measurements and the model are real and which is contributed by the measurement setup, different calibration procedures, etc.

We agree that the spectral uncertainty of the asymmetry factors is not adequately discussed in the text. To correct this, we modified the section 4.1 and added the following discussion at the beginning of the section:

"*Fig. 4 showed that the observed high degree of mesoscopic scale complexity dominates the angular scattering function over the ice crystal shape and a uniform angular scattering function is observed at two wavelengths (532 and 804 nm). Therefore, it is justified to use a single-habit optical ice particle model assuming severely roughened surfaces to compute the bulk optical properties of ice clouds. It was found that the severely roughened column aggregate model showed the best fit of the atmospheric measurements performed at both wavelengths. At 804 nm the model disagreed slightly with the measurements at the sideward angles (Fig. 4). This disagreement indicates that either the severely roughened column aggregate model does not accurately represent the spectral dependence of the asymmetry factors, or could also be related to systematic measurement uncertainties caused by using different measurement systems. However, since we only have information on the ice particle angular scattering properties at two wavelengths at the moment, only one optical particle model is used to parameterize the asymmetry factors.*"

We also changed the last sentence in the first paragraph of the conclusions: *"Moreover, since the ice particle angular scattering functions did not vary significantly between different geographical locations, the modelling efforts of ice particle optical properties in future weather forecast and climate models will be simplified."*

4. A further point about Figure 5 also needs to be noted. Recent theoretical electromagnetic studies have shown that surface roughness, at scattering angles around exact backscatter, induces coherent backscattering, so the phase functions of surface roughened ice should not apparently be flat at exact backscattering angles, there ought to be some backscattering amplitude present. The authors are referred to the following paper for further information about this interesting interference effect, https://www.osapublishing.org/DirectPDFAccess/B8203150-AE8E-68E9- D2CB7062A1AB5EF8_385794/oe-26-10-A508.pdf?da=1&id=385794&seq=0&mobile=no . To compute the phase functions, the authors use a database which probably applies the improved physical optics approximation, in that multiple scattering is not included, so surface roughness is approximated by some geometrical treatment such as facet tilting to smooth the phase functions that appear in Figure 5. As a consequence of this, one could argue that the phase functions presented in Figure 5 are incorrect. Of course, owing to the asymmetry parameter being largely determined by diffraction, its derived value will not be much affected by this backscattering amplitude. However, this still does need to be noted in my opinion to encourage inclusion of multiple scattering in calculating the phase functions,

especially if they are to be used for lidar applications at visible wavelengths. However, to obtain more representative phase functions, the backscattering amplitude could be added on to the phase functions presented in Figure 5. There is a parameterization that the authors could use to do this as explained in this paper https://www.osapublishing.org/oe/abstract.cfm?uri=oe-24-1-620, where IGOM is corrected using the estimated amplitude obtained from electromagnetic calculations.
The Referee points out that recent theoretical electromagnetic studies have shown that surface roughness can lead to coherent backscattering enhancement at angles around exact backscattering whereas the theoretical functions of severely roughened particles showed in Figs. 5 and 6 do not take into consideration this effect. However, for the aspect of energy redistribution in the scattering process the backscattering enhancement has a negligible effect - as also stated by the Referee and, thus the derived asymmetry factor will not be affected through exclusion of this effect. The Referee also pointed out that this effect can have consequences for lidar application, which we agree. Therefore, we added a discussion of this effect to the Chapter 3.4:

“At the angles around exact-backscattering the severely roughened column aggregate model predicts a relatively flat behaviour. However, recent modelling studies have indicated that the scattering intensities around exact backscattering angles should be enhanced due to coherent scattering (e.g. Zhou, 2018). Although this effect can be important for lidar applications, it does not significantly affect the redistribution of the energy in the scattering process and, thus, the magnitude of the asymmetry factor.”

Furthermore, the Referee states that it is arguable that the theoretical phase functions in Figs. 5 and 6 are incorrect due to the treatment of the surface roughness in the model by using the tilted-facet (TF) method. Although the TF method may not accurately represent the physical surface roughness, it has been shown that the TF method can be used to model the phase matrix element P11 with high accuracy (Liu, Panetta and Yang, 2013). Also, arguing whether an optical particle model is incorrect is usually based on comparison of models with more sophisticated models, rarely on a comparison with measurements. This study presents an comparison of one optical data base with atmospheric measurements. In future, it is certainly of interest to perform more such comparisons with also other optical particle models to address the question of which optical models perform the best for different applications.

5. Also, for some reason, the authors do not cite papers prior to 2010, there are some, but these are few and far between and tend to be their own. This needs to be corrected.
We have expanded the list of cited papers. Please refer to answers to minor comments 2, 3, 9, 20 and 21.

Minor comments now follow:

1. In the abstract, the averaged asymmetry parameter of 0.75 is determined at the wavelength of?
We added wavelength after the asymmetry factor.

2. Introduction line 15, similar results by Ulanowski et al., (2006) and Ulanowski et al. 2014 were also reported.
We added citation to Ulanowski et al. (2016) and Ulanowski et al. 2014 to line 15.

3. Introduction line 16, representations of ice crystal surface rough- ness via facet tilting were also added prior to 2008 by Macke et al. (1996)[https://journals.ametsoc.org/doi/pdf/10.1175/1520-0469%281996%29053%3C2813%3ASSPOAI%3E2.0.CO%3B2], Yang and Liou (1998) [Single-scattering properties of complex ice crystals in terrestrial at- mosphere, Contr. Atmos. Phys., 71, 223–248, 1998], Baran et al, (2001)[https://rmets.onlinelibrary.wiley.com/doi/abs/10.1002/qj.49712757711], Baran and Francis (2004)[https://rmets.onlinelibrary.wiley.com/doi/10.1256/qj.03.151], Sun et al. (2004)[https://www.osapublishing.org/ao/abstract.cfm?uri=ao-43-9-1957. There are of course others.
We added both the citations to Make et al. (1996), Yang and Liou (1998), Baran et al. (2001), Baran and Francis (2004) and Sun et al. (2004) and “e.g.” before the citations.

9. Page 2, discussion on polarization, line 2, The

same was also shown by Baran and Labonnote [https://www.sciencedirect.com/science/article/pii/S0022407305003699] in regards to polarization.
We added this reference.

10. Page 3, line 15, replace “in” by “on”.
We corrected this.

11. Page 3, line 25, perhaps, the word “the” needs to be incorporated before “discrete dipole”.
We added “the”.

12. Page 3, line 34, insert the word “to” before “as”. . .
We corrected this.

13. Section 2.2, in the discussion on the PN being used to determine the angular scattering functions, there is no explanation or discussion as to how shattered artefacts were removed from the analysis. Please could you insert this, otherwise, we may be led to believe that those functions could be more pertinent to shattered ice and so will provide low asymmetry parameter estimates.
The Referee is correct that the influence of the shattering artefacts to the PHIPS and PN measurements are not adequately discussed in Section 2.2. Since PHIPS performs particle-by-particle measurements, it is possible to detect shattering events by investigating the particle inter-arrival times. The analysis of the particle inter-arrival times revealed two modes - one mode of short inter arrival times corresponding to shattering events and one mode of longer inter arrival times corresponding to real particle events. These two modes can be separated with a threshold of approximately 1 ms. We have removed all the angular scattering functions identified as shattered events from the analysis.

The analysis of the PN data, including shattering artefacts, is discussed in the original publications cited in this manuscript and in previous studies. For example, the effects of shattering artefacts to the PN measurements are discussed in the Appendix B of Mioche et al. (2017). The authors stated that although it is not possible to avoid or estimate the shattering effects in the PN signal, it can be estimated that the shattering artefacts are within the measurement uncertainty of the PN (25 % on the extinction coefficient).

Two sentences discussing the shattering effects were added to the Section 2.2 after discussion of both instruments:

PHIPS: *“Before analysis, particles corresponding to shattering events were removed by calculating particle inter-arrival times and removing particle pairs with inter-arrival times <1ms.”*

PN: *“It is not possible to correct the PN data for shattering artefacts but it has been estimated that possible shattering artefacts contribute less than 25% to the total extinction signal (Mioche et al., 2017).“*

14. Section 2.4, perhaps save space by compiling the list of campaigns into a table? This improves readability.
Section 2.4 does not only give a list of the campaigns but also discusses the definition “ice cloud” in each of the campaigns (i.e. which cloud types were included in the analysis). The discussion presented in Sect. 2.4 is relevant for understanding the results, since different cloud systems were sampled in different campaigns. Furthermore, this section gives a description, how droplets were excluded from the dataset. For these reasons, we think Sect. 2.4 is important and cannot be reduced as a table.

15. There are many campaigns dating back to before 2010, how did the authors make sure that the PSDs were treated consistently into one database from the variety of differing microphysical probes?
This manuscript reports measurements from three different microphysical probes: the Small Ice Detector 3 (SID-3), the Particle Habit Imaging and Polar Scattering (PHIPS) probe and the Polar Nephelometer (PN). All the SID-3 and PHIPS data from each of the field campaigns are analyzed using the procedures described in the Sections 2.1 and 2.2. Also, the analysis methods of the PN probe have not been modified since published in Gayet et al. 1997.

16. Page 5, line 23, suggest replace “to” with “for” . . . the analysis. . .
We corrected this.

17. Section 2.5, please add a description of the current ice optical parameterization used in ECHAM-HAM. It is often referred to but unknown as to what it actually is.
The current ice optical parameterization in the ECHAM-HAM is calculated with Mie-theory but the asymmetry parameters are scaled down to be more reliable for aspherical ice particles. We added this description of the current optical parameterization to Sect. 4, where the ECHAM-HAM model is discussed.

18. Page 7, line 6, suggest insert the word “to”. . . a change. . ..
We corrected this.

19. Page 7, line 10, comma after aggregates?
We added a comma.

20. Page 8, there are a whole list of studies that predate 2010 in showing that flat featureless phase functions best represent angular short-wave measurements obtained from above ice cloud such as Doutriaux-Boucher et al., (2000)[https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/1999GL010870], Labonnote et al. (2001)[https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2000JD900642]. A more recent paper by Letu et al. (2016) [https://www.atmos-chem- phys.net/16/12287/2016/] uses comprehensive PARASOL short-wave reflectance data to show the same.
We extended the references as suggested.

21. Page 8, line 16, Again, there are many papers that predate 2013, please cite a representative sample.
We extended the references by citing Macke et al. (1996), Yang and Liou (1998), Liou et al. (2000), Baum et al. (2010), Baum et al. (2011), Baran (2012), Diedenhoven et al. (2012). We also added e.g. before the references to illustrate that the cited references are a subsample of literature.

22. Page 9, line 5, typo “sdiscussed”.
We corrected this.

Figures:
Fig. 1 penale-> panel.
We corrected this.

Fig. 2 difficult to distinguish purple from red, suggest changing purple to green.
We changed the purse trajectories to green and modified the colours in Fig. 3 accordingly.

Table 2. Please also insert the percentage of the total particle population rejected owing to shattering.
We have added a new column to the table showing the percentage of ice particles rejected owing to shattering.

References

Gayet, J.-F., Crépel, O., Fournol, J., and Oshchepkov, S.: A new airborne polar Nephelometer for the measurements of optical and micro- physical cloud properties. Part I: Theoretical design, Annales Geophysicae, 15, 451–459, 1997.

Liu C., R. L., Panetta, P., Yang, 2013: The effects of surface roughness on the scattering properties with sizes from the Rayleigh to the geometric-optics regimes. *J. Quant. Spectrosc. Radiat. Transfer.*, 129:169-185.

Mioche, Guillaume, et al. "Vertical distribution of microphysical properties of Arctic springtime low-level mixed-phase clouds over the Greenland and Norwegian seas." *Atmospheric Chemistry and Physics* 17 (2017): 12845-12869.