We thank each of the reviewers for their helpful comments and criticism. We have modified the paper taking into account all of these points and feel that the revised paper is significantly improved and that our results are now presented more clearly and the findings are more robust. Here is a brief overview of the key changes made to the paper.

- **Rewritten paper to be more focussed on the meteorologically interesting aspects of this case.**
- **Improved the clarity of the description of the new retrieval technique**
- **Inclusion of new equations defining the aggregation efficiency and the expected change to the lambda (slope of particle size distribution) with height due to aggregation from Mitchell (1988)**
- **Added more information about the assumptions made, their validity and a comprehensive sensitivity test to these assumptions (new section 6.1)**
- **New figures showing:**
    - **the relationship between diameter and DWR for the Westbrook (2006,2008) scattering model, and for particles from Leinonen and Moisseev (2015) (figure 1)**
    - **Snow flux and number flux as derived from our retrieval (figure 7)**
    - **Sensitivity of the particle size distribution to assumptions in the retrieval (figure 8)**
    - **Sensitivity of the change of lambda (slope of particle size distribution) to assumptions in the retrieval (figure 9)**
    - **Information about the statistical properties of the velocity-diameter power law fits made in the retrieval (figure 10)**

We believe that the improved clarity and additional information now provided in the paper make our results more convincing.

Specific replies to the individual reviewers comments are below.

Reviewer 1

General Comments

The paper focuses on the identification of a rapid aggregation layer within an ice cloud. In order to do so, an innovative algorithm for the retrieval of snow particle size distribution (PSD) is developed. The algorithm leverages on the synergies of multi-frequency and Doppler observations from vertically pointing radar systems. The retrieved evolution of snowflake sizes is connected to microphysical processes through a modeling approach. It is concluded that neither depositional growth nor riming can explain alone the rapid increase in snow size and aggregation must play a major role, moreover, the expected sticking efficiency must be larger than what was previously published in dedicated laboratory experiments.

The paper puts emphasis on the properties of the rapid aggregation layer and in particular to the value of the aggregation efficiency (Eagg). This would entitle the paper to be published on ACP given the importance of this process in ice clouds. However I am not sure if the reported conclusions are sufficiently supported by scientific evidence. In particular, I am concerned about the number of strict assumptions that have been made throughout the text, the lack of independent validation of the results and the very short duration of the single event selected to support the conclusions about Eagg value. By contrast the paper propose a very interesting and innovative way to use vertically pointing radar to retrieve the properties of particle size distributions. As best of my knowledge, this is the first retrieval of the size-resolved PSD using radars, which would allow to drop the assumptions about PSD shape that are necessary in bulk approaches. The presented methodology deserves a much more detailed description than the one presented in the text and a profound analysis of the sensitivity of the method to the various assumptions that have been made. After such revisions, I would definitely recommend to publish it, but I would suggest to consider a different journal such as AMT given the shift of the focus of the paper.

Given all of my concerns, I recommend to consider the paper to be published after a major revision.

We thank the reviewer for their comments, and have ensures that in the revised manuscript there is a clearer description of the retrieval method and a sensitivity analysis to determine the impact of uncertainties in the retrieval. Although the exact details of the particle size distribution showed variability in the sensitivity testing, the conclusion that the broadening of the size distribution is a result of aggregation appears robust. We have therefore decided to keep the paper focussed on the Meteorological aspects of this event and keep the paper within ACP-scope.

Specific comments

1) Given the centrality of the concept for the entire manuscript I would suggest to give a definition of Eagg in the introduction section. This also to prevent potential confusion, given by the non-unique nomenclature used in this field where different efficiencies might mean different things (e.g. collection efficiency, sticking efficiency). Finally, this would help understanding the reasoning behind the last paragraph of section 6 and Figure 5, where Eagg is inferred from the vertical gradient of the slope parameter of the PSD.

Thank you for the suggestion. We have added the formal definition of aggregation efficiency based on Mitchell (1988)'s aggregation kernel to the paper – see equation 1 and the supporting text.

2) I am not convinced by the statement about Connolly et al. (2012) at lines 31-34 of page 2. By looking at Fig. 14 in the original paper I would agree on the fact that Eagg is between 0.4 and 0.9 at -15 C because that is the confidence interval reported in the plot. For the very same reason I would say that it is between 0 and 0.5 for the other temperatures. Claiming that it is always below 0.2 might be an exaggerated statement.

We have revised the text to clarify that the "best estimate" of E_agg is below 0.2, except at -15C.

3) In section 2.1 the non-microphysical sources of differential reflectivity (DWR) are accounted for. These sources of retrieval error are compensated by making the radar reflectivity to match in the Rayleigh-scattering part of the cloud and applying the same adjustment to the whole profile (lines 4-5 of page 4). This method is strictly valid only for radar miscalibration and attenuation by radome or wet antenna; for height dependent sources of differential attenuation (e.g. atmospheric gas absorption, liquid in the cloud) this method causes an overcompensation of the higher frequency reflectivities at lower level (in particular W-band). Attenuation due to atmospheric gases depends on the density and humidity profile of the atmosphere and can adds up to 2.5 dB at the top of the cloud at midlatitude; under this condition the overcompensation caused by the radar cross-calibration at 3-4 km could be easily in the order of 1 dB. I suggest to compensate for atmospheric gases absorption profile before making the radar-cross calibration, or, at least, estimate the W-band absorption profile for the analyzed case by using either a weather model or a radiosonde profile and report it either in the paper or in supplementary material.

We understand the reviewers comment and have considered this issue. However, it should be noted that the overwhelming majority of attenuation from liquid water and gases (and therefore also differential attenuation) occurs in the lower troposphere, below the level of clouds we are analysing in this paper. Therefore, our application is analogous to that of radome attenuation (where the attenuation occurs before the first target of interest). To further support this argument, we have calculated the attenuation from water vapor as a function of height (using a nearby radiosonde profile), and determined that 85% of the attenuation occurs below cloud base. The remaining attenuation would contribute to about 0.15 dB difference of 35 and 94 GHz power. Such an offset in power translates to about 60 microns in difference of the retrieved particle size at both cloud base and cloud top. Therefore, the conclusions drawn about the rapid aggregation occurring in this cloud are not affected by the attenuation correction we have made. However, the reviewer is correct to point out that there will be some situations where this method will not work correctly and we have added a sentence to the paper stating this to discourage future researchers from blindly applying this method.

Additionally, the fact that the spectra at all 3 frequencies overlay one another nicely in the upper levels of the cloud suggest the relative calibration works well. If there were significant non-Rayleigh effects, they would be more prominent in the spectra which would show sZ94 too high for small (slow falling) particles, and sZ94 too low for large (fast falling particles). We don't see that behaviour. In order to address the reviewers concerns, we have added different possible definitions of the "Rayleigh-scattering regime" in which we match the reflectivity from the 3 radars as part of the sensitivity testing. Our findings appear robust to which range of dBZ values are chosen.

4) 35- and 94- GHz radars are declared to be off-zenith by 0.2 and 0.15 deg in opposite directions (lines 15-16 of page 4). This causes a contamination of the doppler signal from horizontal wind component which is then corrected by shifting the spectra by constant values (line 2 page 5). The authors acknowledge the fact that this procedure is imperfect and consider the matching of the resulting spectra to be good in Figure 4. However it is not described how these numbers (mispointing angle and Doppler velocity correction) are obtained. I personally can hardly see how a composite relative shift of just 0.1 m/s would have affected the matching in Figure 4. The comparison in Figure 4 would have taken benefit from the inclusion of the 3 GHz spectrum which is considered to be well aligned. Also the 'goodness' of the matching is both affected by velocity and power shifts: a small residual differential attenuation would have caused the spectra to look non-aligned; given the fact that there might be still a differential attenuation issue here (see my point number 3) the matching is potentially flawed by this residual error.

I suggest to include the source of the mispointing angles and Doppler shifts numbers.

Following the reviewers suggestion, we have also added the 3 GHz spectra to the plots in Figure 4 (now Figure 5). In accordance to our reply to the previous point, the goodness of fit of all three spectra (in regions where we expect them to be identical, e.g. figure 4a (now figure5a)) provided by making these corrections to the measured Doppler velocity suggest that it is necessary and beneficial. Contrary to the reviewers expectations, a mis-alignment of the spectra by even 0.02 m/s can show substantial differences in the retrieved particle size distribution. This difference is because the spectra is relatively "steep", sZ changing rapidly as a function of velocity. Hence, slightly misaligned spectra result in artificially enhanced sDWR values, either for small velocities, or for large velocities. A relative offset of 0.1 m/s renders the retrieval useless – see the retrieved size spectra when such an uncertainty is added in the new sensitivity analysis. The importance of the velocity offset for the retrieval has been clarified in the revised paper.

A description of how the offsets and pointing angle errors were calculated was not included in the original paper because the cause of the offsets is not of practical significance, but the fact that we have corrected the data to account for them is important. For completeness we have added a short description to the paper and also here:

The velocity differences as a function of height were determined be assessing the mean Doppler velocities from the three radars individually. The correlation of these offsets with the atmospheric wind profile (determined from ECMWF forecast fields) enabled an estimation of the pointing angle errors. We include these values in the paper only to highlight to future researchers that rather small pointing angle errors can significantly affect the retrievals and therefore care should be taken to ensure that the radars are pointed as accurately as possible.

The goodness of matching of the spectra is indeed affected by both velocity and power offsets. However, these are easy to separate and correct for independently. The velocity offset can be determined through correlation of the Doppler spectra while power offsets can be determined by integrating received power across the spectra. In fact, we used bulk methods to determine the offsets (mean velocity difference between radars and total reflectivity differences – as discussed above) and these also worked well to ensure the Doppler spectra are well matched.

5) The PSD and v-D retrieval method is very roughly explained in pages 6 and 7. After several readings I understood that it assumed a unique relation between sDWR and the snowflake size. This relation is likely to be very specific of the assumed scattering model and mass-size relation. A plot showing this relation for a certain number of scattering models and m-D functions will help the reader understanding the methodology applied and gives an indication of the expected uncertainty due to the related assumptions.

The reviewer is correct that this section was not sufficiently clear in the submitted version. The text has been improved to add clarity and the figure suggested has been added to the paper (new Figure 1) to highlight both the method and the characteristics of the scattering model used.

6) Moreover, for the scattering model it is assumed Westbrook (2006, 2008a) since it has been found to closely match observation in the multifrequency space [Stein 2015]. However, the scattering model from Leinonen and Szyrmer (2015) has also been found to match the observation (unpublished on a peer review journal, but included in conference proceedings http://www.isac.cnr.it/ ∼ ipwg/meetings/bologna2016/Bologna2016_Orals/3-8_Westbrook.pdf) It would be very interesting to see the results from this different scattering model. Being a detailed DDA model one does not have to assume the m-D relation but can simply take particles masses and sizes from the database, achieving a better consistency of the results.

While comparison with other scattering models is indeed interesting, we present plenty of evidence in this paper, and also in Stein et al. 2015, that this scattering model is suitable for this case. The Leinonen and Szyrmer (2015) scattering model is for rimed aggregates, and we have already stated in the paper that there is no evidence for riming, either in terms of the presence of supercooled liquid water or the characteristic behaviour of the pair of DWR values presented in Stein et al (2015). However, the scattering calculations in the presentation that you link to are indeed for unrimed aggregates – these have very similar characteristics to the Westbrook (2006) scattering model used. We have added points to Figure 1 to highlight the similarities of the two scattering models.

Additionally, it should be noted that we do not require a mass-size relationship to relate sDWR and particle size – the retrieval method for size is independent of the mass-size relationship.

7) The particles that are sampled within each Doppler bin are likely to have different sizes. Is the model considering only one particle size per Doppler bin? This is potentially a significant source of error when large particles are present. Large particles are expected to fall roughly at the same velocity for many different sizes, thus dv/dD « 1, by contrast the backscattering signal given by those particles is very different. Assuming that in fast-falling doppler bins (i.e. v>1m/s) there are snow particles of just one size is not a valid assumption even at for doppler systems with a very high spectral resolution.

We agree with the reviewer that the relationship between particle size and particle velocity is likely not a one-to-one monotonic function. This assumption and the limitations have been further discussed in the revised paper compared to the original version. However, as a first attempt at using such a technique to retrieve the particle size distribution without fixed assumptions about the shape of the distribution - we need to make some assumption here. It may well be that a refined approach could yield more accurate or robust results; however, this would require a-priori information on the statistical variability in V for a given D, which is poorly constrained at present, and therefore we leave that for future work.

8) Considering the number of correction, a sensitivity analysis of the algorithm with respect to the input data is essential. Assuming 1 dB uncertainty in radar reflectivity and 1 or 2 velocity bins uncertainty in the doppler spectra will already give a good indication of the robustness of the algorithm. It will be particularly interesting to see how this translates into uncertainties in the retrieved PSDs (panels c, f and i of figure 4) and the profile of fitted scale parameter lambda in figure 5.

A sensitivity analysis incorporating uncertainties of +/- 1 dB to the Doppler spectra, a shift in velocity space of up to +/- 0.04 m/s, which range of reflectivity values are used for attenuation correction and an additional mass-diameter relationship has been added. The impact of the uncertainties of the size distribution (equivalent to figure 4i) and of the vertical profile of Lambda (equivalent to figure 5) have been added. The sensitivity analysis adds confidence to our argument that aggregation is the primary driver of change to the size distribution in the lower region of the cloud. Furthermore, it shows the (negative) impact on the particle size distribution retrieval when the Doppler spectra are not suitable matched – suggesting that our matching methodology is in fact reliable (albeit imperfect).

9) The result of 'rapid aggregation' is obtained by comparing the relative potential of various snow growth processes, concluding that only aggregation is capable to give such rapid change in PSDs scale parameter lambda. I think that the potential given by the PSDs retrieval is here underutilized. Given the full PSD and the m-D and v-D relations one can calculate interesting bulk quantities such as particle number concentration (Nt) and ice water content (IWC) and their vertical fluxes (particle flux Nf and snow rate SR). It will be extremely interesting to see time-height plots of this quantities in connection with the results in figure 4 and 5. For instance, positive variation of Nf and SR should be seen in connection with the newly developed mode in fig4d. This analysis would also help in the identification of the significant aggregation process. Depositional growth and riming are in fact expected to increase SR leaving Nf unchanged (unless newly nucleating particles are present). Aggregation has the distinctive effect of decreasing Nf leaving SR unchanged and this should appear in the suggested plots.

We thank the reviewer for this suggestion, which has resulted in the addition of a new figure showing the number flux and snow flux throughout the interesting part of the observed cloud. We believe that the coherence in these plots adds support that our retrieval is stable and as the revierer suggested – the vertical profile of the number and snow flux do add support to the rapid

aggregation hypothesis. Although the snow flux decreases with height (presumably due to sublimation), the number flux decreases with height significantly faster.

10) At line 27 of page 12 it is mentioned that the methodology described in Mitchell (1988) has been used to model the evolution of lambda parameter, however it is not specified the exact model used. It is surprising that the formula for the depositional growth rate from Pruppacher and Klett (1978) has been fully reported and not this. This is potentially a serious issue regarding the reproducibility of the results. Additionally, I think that a better explanation of the model used will give the reader a better understanding of the other variables that influence the PSD evolution due to aggregation such as particle sizes, velocity differences and total number concentration.

We apologise for this oversight and have now added the full equation from Mitchell (1988) to our paper. Furthermore, because of the additional analysis of the snow flux added, we decided that the constant snow-flux with height assumption was not valid, and instead used the retrieved snow flux profile in the calculation. We have maintained the constant snow-flux profile for E_agg of 1.0 in the paper for comparison.

11) The conclusions about the value of the Eagg are supported by only a 2 minute average profile during one event. I would, at least, model the evolution of the lambda parameter for other times during the same event, or, even better, model more events.

While we understand the reviewers request for the analysis of more data, we focus in this paper on the most microphysically interesting part of the cloud. We do not claim to make any general quantification of the aggregation efficiency, but rather to say that in this instance the observations are consistent with a large aggregation efficiency and the new retrieval has helped identify this process.

We have collected radar data from several days and believe that we have seen similar events within that dataset. We have focussed on this case study as it is microphysically interesting enough to serve as an example of the new retrieval technique and the insights into cloud microphysical processes that it can provide. The analysis of further events, where similar features were observed, are underway. The analysis of these events is not ready to be included in this paper and will hopefully be published separately (although the research grant for this work has now expired).

Technical corrections

12) When presenting the state of the art of Doppler/multi-frequency radar retrievals at line 20-25 of page 2, I suggest to consider some recent studies like Chase et al. (2018) or Leinonen et al. (2018) in the discussion.

Thanks. These more recent papers have been added to this discussion.

13) The choice of the colormap used in figures 2, 3 and 5 is particularly unfortunate. There is an apparent overlap of light-blue colors for different values that makes the interpretation of the figures more difficult than it should be. In figure 4b there are vast areas of the cloud where I cannot say if the DWR is either +1 or -1 dB. In figure 5 the mapping from the colorplot to the profile is made even more difficult by the fact that the profile as been cut from the panel with a white line; here I also suggest to indicate the profile with a thin rectangle around instead of the white line.

Actually the white line in figure 5 is because of missing data. The profile is at 1615 UTC, which has been emphasised by adding the time to the title of panel c and as an additional label on the x-axis of panels a and b.

We have experimented with different colour scales; however, none are able to cover the full data range while enabling values to be read from the figure accurately. It is because of this that we added vertical profiles of the values at 1615 UTC to figures 3, 6 and 7.

14) Figure 4 – Personally I would swap the axis in panels b, e, h. This would put velocity on x-axis, matching the concept on panels a, d and g. Also it appears that DWR is rather a function of velocity and not the opposite (see in particular panel e). That is a personal preference and I would leave to the authors the decision.

This is a good suggestion and has been changed in the new version of the figure.

Reviewer2

1 Summary

This manuscript proposes a new algorithm to retrieve the particle size distribution (PSD) from vertically pointing Doppler profilers at 3 frequencies, using the spectral dual-wavelength ratio and not the ratio of integrated reflectivity values as in previous work. This algorithm is then applied in the context of the study of a given cloud, to investigate the dominant microphysical processes taking place and explaining the measured Doppler spectra. Rapid aggregation appears to be the best candidate among various processes to explain the observed behavior.

2 Recommendation

The algorithm and the application for microphysical interpretation that are presented in this manuscript are innovative and relevant. A “direct" PSD estimation without any assumption about its mathematical functional form is promising and useful. But there are also a number of assumptions that are required to run this “inversion", and they are not all clearly described and discussed. It is hence difficult to understand in which framework this approach can be safely used, and the example presented in this manuscript remains rather specific. The manuscript is pleasant to read with quality illustrations. Overall, I am convinced that this manuscript presents innovative and original material that are worth publication, but after having addressed the issues listed below.

We thank the reviewer for their supportive comments. We have developed the manuscript in response to the reviewers comments (as detailed below) with a more rigorous analysis of the uncertainties and a better description of the assumptions and retrieval technique. As a result, we feel this has made the paper much more convincing and the results robust.

3 General comments

1. Information about the methodological side is missing: no detailed/exhaustive description of the proposed PSD spectral retrieval algorithm is provided, making it difficult to check or reproduce for instance. I suggest the authors to add detailed description (including equations and so on) of the different steps of the algorithm.

The algorithm is actually relatively simple, but we acknowledge that the description could have made the process clearer to the reader. We have added additional text and clarifications and believe that the revised manuscript provides a better basis for reproducing this method.

2. The case study is too limited (40 min of a single cloud) to derive general insights beyond the demonstration that the proposed method works, at least for one cloud. I understand the difficulty to expand the analysis, but this example is too limited in itself (see below).

The focus on this case is because of the interesting, abrupt in height but consistent in time, appearance of large DWR values lower in the cloud. We do not try to claim that this is a common feature, or that the aggregation efficiencies derived are common to other clouds/cloud types. As discussed in the paper, data on the aggregation efficiency is rare and there is a large spread in the reported values. Therefore we believe the additional insight from this paper to be valuable to the community. We present an analysis of what is occurring at this time and height in this cloud, and show how this method can be useful to investigate processes in such clouds. Further study of other clouds using the same method is underway at a preliminary stage, and we hope that this study and retrieval technique will provide a foundation to analyse the variability of aggregation

efficiency in clouds in a systematic way and to evaluate how it depends on temperature, relative humidity etc.

3. From a more general point of view, I have the feeling that this manuscript “oscillates" between the two Copernicus journals AMT and ACP, between a more methodological point of view (e.g. the retrieval algorithm) and a more meteorological point of view (case study of rapid aggregation in a cloud). So in the end, the reader is somehow frustrated: on the one hand, the paper proposes a new retrieval method (AMT side), but does not provide enough description of this method for the reader to implement it; on the other hand the case study is too limited to gain any general insights into cloud microphysics (ACP side). I am fine with the authors choosing ACP, but I would strongly recommend to add more explanations about the proposed retrieval technique, as well as more discussion about the limitations and the conditions in which this approach is valid.

There is some content in this direction in the conclusion (p.15, l.7-11) but only the verticality and the beam width are discussed, not the requirements in terms of turbulence, (supercooled-)liquid water or not, the geographical representativity, etc.

Based on this comment and that from other reviewers, we have tried to focus more on the meteorological aspects within the paper, but at the same time clarifying adding some more details about the retrieval technique.

4 Specific comments

1. P.8, l.2: optimal with respect to what? Which fitting method is employed to estimate the power-law parameters?

The word optimal has been removed. The power-law was estimated by fitting a linear best-fit line to the logarithm of the values.

2. P.8, l.2: why using a power law between vertical terminal velocity and the size?

The power law has been used because 1) it is easily differentiable and 2) it is common in microphysical scaling relationships.

3. P.8, l.10: so the 3 GHz spectra are used “only" for large particles? If so, the proposed approach is essentially dual-frequency. Should the title be adapted?

The 3-GHz is essential for the attenuation correction of the radars (because it is used to identify the Rayleigh-scattering part of the cloud and provide a reference). It is used in the sizing of particles larger than 2.2 mm (which can be done for both 35 and 94 GHz, and should provide the same answer). It is furthermore useful to help identify the correct scattering model to use – as done in Stein et al (2015). Therefore, although some aspects only employ dual-frequency techniques, the complete retrieval is dependent on all three radars. The 3GHz spectra has additionally been added to Figure 5 to enable comparison between all three radars.

4. P.10, l.26-27: what are the plausible mechanisms to explain the generation of these new ice particles? Maybe it was mentioned somewhere but if so, I missed it.

We have not speculated on the generation mechanism because we have no data that will help determine or rule out any mechanism.

5. P.11, l.25-27: is a SNR threshold applied prior to run the retrieval, in order to filter out the noisy values?

Yes, noisy data points are filtered out and details have been added to the text.

Reviewer 3

The authors present a method to quantify the aggregation process and retrieve the ice particle size distribution using three co-located radars. They showed that aggregation causes a rapid (less than 10 minutes) growth of ice particles from 0.75 mm to 5 mm in maximum size. They speculate that the dendrites dominate at -15 C with large aggregation efficiency (approximated to be near unity). Although the results are important and the manuscript is interesting, there are multiple issues that have to be addressed before the manuscript can be accepted for publication. My suggestions are explained below.

General comments:

- How do you distinguish between the ice particles and water drops? In pg 5, ln 21, you said that your case is an ice cloud. Elsewhere you mentioned that there was no water drop in the cloud. However, a mixed-phase cloud is probable in this temperature range. Fig. 3 shows that the temperature in the presence of cloud ranges from 0 to -40 C. Between -38 and 0 C, super-cooled water drops co-exist with ice particles (Rosenfeld and Woodley, 2000), and there is a great chance of water contamination.

It is important to address this, and explain how you detect water drops and exclude them. Alternatively, is it possible to quantify the ratio of liquid water content to ice water content?

Mixed-phase clouds are possible in this temperature range; however, we are confident that the liquid water content in this cloud is negligible. Firstly, the microwave radiometer instrument does not detect any significant liquid water. Second, analysis of the radar Doppler spectra does not show any evidence of low reflectivity drops at small fall velocities (although they could be too small to be detectable). Third, the evidence of the pairs of DWR-values shown in Stein et al. (2015) are consistent with aggregates, and inconsistent with rimed particles – which suggests that there isn't a lot of supercooled water present. These clarifications are all included in the revised paper.

- There is no comparison between your retrieval and direct measurements of size spectra, because there was no in-situ measurement available for your case. It is true that disagreements exist between various in-situ probes (see also Fig. 6 in Cotton et al., 2010), but still it is not certain if your retrieved size spectra would be more accurate. It would be good to cite any study that compared retrieved size spectra with direct observations. In any case, such caveat (no comparison between your retrieval and in-situ measurements) should be explained in the manuscript, and should be mentioned in the abstract and conclusions.

We do not try to argue that our method is necessarily more accurate than in-situ measurements. However, the advantage is that we can make continuous measurements and multiple heights simultaneously and see the evolution of the size distribution. It is unfortunate that there is no in-situ data available, and such a comparison is part of the planned future work.

- The Brown and Francis (1995) mass-size relation has an important issue: it's not realistic for size smaller than 100 microns, since it gives ice particle mass larger than that of a sphere. See Erfani and Mitchell (2016) and their Fig. 1. I understand that you do not detect particle smaller than 0.75 mm, but it is important to address this issue for the readers who might use Brown and Francis mass-size relation. In addition, the readers will become aware of the more recent mass-size relations.

We agree that the Brown and Francis (1995) mass-diameter relationship is not physical for small sizes. However, such a failing does not affect our retrieval because it is not possible to reliably size

any particles smaller than 0.75 mm, and certainly not down to the 100 micron scale. We also note that Brown and Francis do address this issue in their original paper and for these reasons it seems unnecessary to include a repetition of that information in our paper.

As part of the newly added sensitivity analysis, uncertainty to the mass-diameter relationship is estimated. We also have evidence from the Stein et al. (2015) paper that the exponent (1.9) used in the Brown and Francis mass-diameter is consistent with the observations on this day.

- Your radar is unable to detect particles smaller than 0.75 mm. This means that your retrieved data is not able to approximate the vast majority of particle number density or dN/dD (because small particles dominate the number concentration or N; again see Fig. 6 in Cotton et al., 2010). How does that affect your calculations? Since the calculation of number concentration is an important part of your paper, you should highlight this limitation (no detection for size less than 0.75 mm) and its consequences in the abstract and conclusions.

Perhaps it was not clearly written in the paper, but it is incorrect to say that the radars cannot detect particles smaller than 0.75 mm diameter, but rather that their size cannot be determined reliably. We have attempted to clarify any possible misunderstanding in the revised paper, including a statement in the abstract that the size distribution can only be estimated for particles >0.75mm in diameter. The radars detect all particles of all sizes (assuming that there is enough total signal to differentiate that from background noise). While you are correct that the retrieval of the number concentration is important for the size distribution – we only attempt to retrieve the size distribution where sufficiently large particles exist. We could extrapolate back to small sizes from the fitted size distribution to determine an approximate number of small particles, but we have no need to do this for our study.

Specific comments:

- abstract, ln 5: Did you calculate the mean size change by aggregation?

Note that we are not referring to the mean size here (an advantage of our spectral technique). We argue that almost all of the size change is due to aggregation. We explain in the next sentence in the paper that the increase in size is shown to be consistent with aggregation when Eagg=0.7.

- abstract, ln 11: Any evidence to support this? I understand that this is suggested based on previous studies. If yes, it should be mentioned explicitly: "Based on previous study, we suggest ..."

There is no direct evidence of this; however, such a process would be consistent with large aggregation efficiencies.

- pg 2, ln 7: By "cloud microphysical properties", do you mean individual ice particle properties such particle size or mass?

Yes, but not only individual ice particles properties, also bulk properties such as ice water content or equivalent properties for liquid water.

- pg 2, ln 14: Please add at least one example (with a reference) on how different size spectra affect the relative importance of microphysical processes.

We have added examples of why vapor deposition, riming and aggregation are affected by the particle size distribution. Details have been added to the text. Furthermore we have added numerous references to the importance of particle size distributions on correctly simulating different cloud types (page 3, line 19-25).

- pg 2, ln 15: Please add a reference.

The sentence reads "Another important application is to provide observations with which numerical models can be evaluated and their parameterizations improved." We do not think any reference is necessary here.

- pg 2, last paragraph: It is good to cite Keith and Saunders (1989), since they performed experiments and measured the aggregation efficiency for various shapes and sizes. They showed that the aggregation efficiency ranges between 0.3 and 0.85 for planar snow crystals depending on the particle size.

Thanks, we were previously unaware of this paper and we have now added this reference to the discussion

- pg 3, Section 2: Please add proper references for each radar and for the near-field correction method. Overall, this section doesn't have sufficient citations and I can see only 2 references in the whole section.

We have added the references for the three radars. The near-field correction was derived empirically by comparison of the 3 and 35 GHz radar profiles in low-reflectivity (Rayleigh scattering) ice clouds and is discussed briefly in Stein et al. (2015). This section is describing the data that we collected, and therefore additional citations would not be relevant here.

- pg 4, paragraph starting at ln 14: Have you tried to correct the direction of 2 radars and make a few measurements, and then compare with the previous measurements?

The errors in the pointing angles were only identified through the analysis in this paper. We could therefore not correct the pointing angles in time to observe this cloud.

- Table 1: Right now, it is not mentioned anywhere in the manuscript.

This has now been corrected.

- pg 5, ln 26: Are these temperatures measured by radiosonde?

The temperatures were from a nearby radiosonde and also from ECMWF forecasts.

- pg 5, ln 28, Change to "Figure 2b".

The reference to "Figure 2" is correct since we refer to both the time series of reflectivity and DWR, which are shown in separate panels.

- pg 5, ln 30: Was the Westbrook model initialized for the same cloud?

The scattering model predicts the observed radar reflectivity based on characteristics of the ice aggregates generated by an idealised model of the aggregation process. It is therefore a statistical relationship and does not use any measurements of the cloud on this day. However, as shown by Stein et al. (2015), there is good agreement between the expected behaviour of the DWR pairs and that predicted by the Westbrook scattering model.

- Fig. 2b an 2c: The explanation of Fig. 2b in the manuscript is not enough. What is the physical interpretation of such difference between the two radars. Also, the explanation of Fig. 2c is missing in the manuscript.

Further explanation has been added of panels a and b, explaining the quantities further and adding brief physical interpretation. Panel c is now explicitly referenced within this discussion.

- Fig.3 and 4: You explained Fig. 4 in the manuscript earlier than Fig. 3, so please switch these figures.

The discussion of figure 3 has been moved earlier

- pg 6, ln 6: Briefly define the scattering model. Also, do you mean individual ice particle or a bulk property such as mean size or median size?

The discussion has been complimented by the new Figure 1 showing how the DWR values change as a function of particle diameter for the scattering model. The diameter is that of an individual particle.

- pg 7, ln 2: See the general comment regarding mass-size relation. It would be good to cite Erfani and Mitchell (2016) since they explained recent mass-size relations.

The general comment was noted. As a result of reviewers comments, the sensitivity to the choice of mass-diameter relationship is now explicitly included within the sensitivity testing. Erfani and Mitchell (2016) provide more accurate but more complex mass-diameter relationships, whereas the estimation of the aggregation efficiency from Mitchell (1988) requires a mass-diameter relationship of the form m=aD^b.

- pg 7, ln 1-4: The steps 2 and 3 need to include the relationships you used to relate various variables.

We have clarified this section to address these and other concerns.

- Fig. 4: When the x-axis says “particle diameter”, do you mean the maximum size of each particle, or did you calculate the sphere-equivalent diameter? Moreover, the explanation of panels b-e-h is missing in the manuscript.

The particle diameter is the maximum dimension, following Westbrook et al. (2006) Discussion of panels b-e-h added.

- pg 10, ln 15-16: When particle sizes grow, but their fall speed does not increases, this is a sign of branching and aggregation rather than riming. See Locatelli and Hobbs (1974).

At this point in the manuscript we do not make any assertions about the processes involved.

- pg 10, ln 17-24: Combine all these lines into one paragraph.

Done

- pg 10, ln 32: Doppler spectra is not bi-modal in Fig. 4a. Do you mean Fig. 4b?

Yes, corrected. Thanks.

- pg 10, ln 33: Why are such small particles a result of nucleation and not growth by vapor deposition, or a secondary ice production (such as fragmentation of ice particles)? Elsewhere you assumed the small particles in the bi-modal spectra are the result of vapor deposition. Any evidence on the mechanism responsible for the increase in small particles?

You are correct, we do not know how these particles have formed, possibly through nucleation, shattering or other processes. Nucleated has been replaced by formed in the text.

- pg 11, ln 1: You say the aggregation causes ice particles to grow larger and fall faster, but aggregate fall speed does not grow by size. See Locatelli and Hobbs (1974) and their Fig. 12. They also provide fall speed-size relations for various ice particle shapes (including dendrites and aggregates). It's good to cite this paper, and also it would be great if you fit their relation to your data and calculate the R-squared.

This is not what the sentence states. It says that the sDWR increases for the larger particles (which are also the fastest falling particles), but not that they necessarily fall faster.

- pg 11, ln 3-4: This can be a sign of aggregation.

This is the argument that we make later in the paper. Here we only present the evidence.

- pg 11, ln 11: This is an exponential function. Moreover, I assume D and dN/dD are known in this equation. How did you calculate N0? It is important to explain this in the paper. It seems that the value of slope is dependent on the calculation of N0. Furthermore, do you use such distribution to relate size to radar reflectivity? Your size spectra do not include small particles. Since the number of small particles contributes significantly to the number concentration, how did this affect your calculations?

Both N0 and Lambda are determined by fitting a straight line to the size distribution data (D and log10 (dN/dD)).
Such a fixed size distribution shape is not part of the retrieval, which is one advantage of our method.

- pg 11, ln 13: This is a qualitative comparison. Have you looked at the difference between Fig. 5a and 5b? From Fig. 5c, it seems the agreement between the 2 slopes is not excellent. Note that this is a logarithmic axis and I can see the red line can be larger by a factor of 1.5.

No, we have not performed a quantitative comparison. However, there good agreement in the values and patterns shown by the two methods. The disagreement at lower heights possibly highlights a weakness in the DWR method, where a fixed size distribution shape is required. We have no expectation that these two (nearly-independent) methods would give such a good agreement on the trend of lambda with height. The absolute values are not particularly interesting for our purposes.

- pg 12, ln 10-12: How did you calculate F (ventilation coefficient) and K (thermal conductivity)?

Suitable values for the appropriate temperature range were chosen.
(K=0.024, Rogers & Yau, chapter 7;
F=0.65 + 0.44*0.6^0.33 Re^0.5;
Re = rho * D * V(D) / dynamic_viscosity;
dynamic_viscosity is in the range 1.512E-5 … 1.862E-5, depending on temperature.
A clearer description has been added to the text.

- pg 12, ln 26-27: The vapor deposition and riming do not change the total number of ice particles (N), but they do change the number of ice particles within each size bin (dN/dD). Please clarify that the rate of change in size is not the same for all sizes. As an example for riming: riming collision efficiency is a strong function of ice particle size, so larger ice particles would grow faster due to riming. See Wang and Ji (2000) and their Fig. 7; might be good to cite this paper.

We have chosen to remove this statement from the paper as it is not key to the argument that aggregation is the main process acting in this cloud and adds unnecessary confusion. We present

sufficient evidence in the rest of the paper that aggregation is important and that deposition and riming are not the key processes involved.

- pg 12, ln 27-28: Please refer to the proper equation number in Mitchell (1988).

The full equation is now reproduced in the paper.

- pg 14, last paragraph: This statement is suited for the Introduction and can be moved near the end of Introduction as the motivation for your study.

Agreed. This paragraph of text has been moved

- pg 15, first paragraph: See my general comments on the lack of comparison with in-situ measurements; I agree the issues exists in directly measuring the particle size and concentration, but still it is unclear how your method has better accuracy. In addition, please cite Cotton et al. (2010) when explaining the disagreements in the in-situ measurements of ice particles.

Again, see the above comment. We do not - in general - claim to be more accurate than in-situ retrievals. We would like to compare our results to in-situ sampling.

Reviewer Paul Connolly

This is a very well put together study, combining data from three co-located, vertically pointing radars to quantify aggregation efficiencies in the atmosphere. It is the first attempt to retrieve the ice particle size distribution from multi-frequency Doppler spectra observations. It is satisfying to see that these results, in the main, corroborate our chamber observations. The presentation is very good and there are no major issues. I recommend publication, but would like to see more information on the fall speed relations used and perhaps an assessment of how the results depend on mass - dimension relations.

We thank the reviewer for the positive reception and interest in our results, and additionally for trying to recompute some parts of our analysis as it was helpful in highlighting which bits of information were missing from the paper.

As I have worked on similar problems before I wanted to see whether I could reproduce the findings from the information available in the paper, to check that my interpretation is consistent with the main findings in the paper. My reasons for doing this are to demonstrate how others may interpret your data, and to check that my interpretation is correct. I present this alternate analysis, in a separate section below.

Specific Comments

Page 3, line 25 – sentence begins with “because”.

Correct. Because the subordinate clause is followed directly by the main clause, this sentence is grammatically correct.

Page 7: Brown and Francis to convert between mass and size . The Brown and Francis (1995) relation is for ice crystals in cirrus clouds. There are more up to date mass - size relations that are published so I was curious if you have tried these, and whether a different assumption affects the results

This is a useful suggestion. In the newly presented sensitivity analysis, a second mass-diameter relationship has been used to evaluate its importance (it turns out not to be very important). Additionally, it should be noted that the mass-diameter relationship plays no role in determining the size of the particle from the measured sDWR (only the number), and that the exponent in the mass-size relationship is shown to be consistent with the Brown-Francis value (1.9) for this case based on the fractal dimension calculation of Stein et al. (2015).

Page 8: velocity power law – you don’t give examples of the fit parameters here, which makes it more difficult for others to understand your data. I wonder if you could give some example figures, or statistics of the fit parameters (the a and b coefficients).

Thanks for the idea. We calculated the mean and standard deviations of the a and b values (in our paper called c and d), and included these in figure 10.

Figure 4: convincing plot. Just a comment: I am surprised that the spectral reflectivity of the middle plot and bottom plot extends to just above 1.5 m/s , whereas the size distributions are much broader for the lower layer. Is this because the larger particles in the lower layer are less dense so that their fall speed saturates with increasing particle size?

Yes, this is an interesting aspect of this case, and already partly commented on in the text – that the sDWR of the particles around 1.5 m/s increases, but the fall velocity doesn’t really change. As

you suggest, this could be explained by a change in density, or an insensitivity of the fall velocity to aggregation at this particular size. We do not have any data to do anything other than speculate about this.

Alternate Analysis

Without responding point by point to your alternate analysis , we wanted to thank you for this analysis as it 1) helped highlight which parts were missing in the paper to enable it to be understood and reproduceable and 2) brought to our attention the importance of the I_1 term in the Mitchell (1988) equation, as well as the mass-size and velocity-size assumptions in that calculation. We have taken your comments on board and revised the manuscript adding the relevant details. This has given us more confidence that our results are robust and has improved the quality of the paper and made our arguments more convincing. A short summary of these is below:

- **In terms of calculating the mass flux, the velocity-size relationship is not used. Instead the Doppler velocity from the radar is used directly (details added to the paper). The snow flux and number flux (for particles D>0.75mm) is now shown in Figure 7. The values you estimated were close to ours and can be seen in the profile in Figure 7c.**
- **The velocity-size relationship is an important contributor to the I_1 term of Mitchell (1988). Your analysis revealed that I_1 is very sensitive to the velocity-size relationship used, and consequently a large uncertainty in the estimated aggregation efficiency evident. This has been brought out in the discussion in the text of the paper.**
- **Statistics of the velocity-size relationship power law fit performed have been calculated and added to the paper in the form of figure 10 and additional discussion has been added to the end of section 6 where we have through in detail about the sensitivity of our results to the various parameters input to the equation and their importance (e.g. a, b, c, d).**

Validation : in order to better understand figure 4 I thought I would do a consistency check. I digitised your data from plots in figure 4 c , f , and i First I wanted to calculate the mass flux, to see if this was roughly in - line with that expected and to see whether it was approximately conserved between levels. As you are aware the mass flux should be conserved in diffusional growth is not important. I used the Brown and Francis (1995) relation to convert particle diameter to mass (as you have done)

And, as you have not given the coefficients for the velocity size relation, I have used a fall speed relation from Wang and Chang (1993)

My analysis is shown in Figure 1 . I have calculated the mass flux at the top, middle, and bottom of the cloud presented in your figure 4 . The values I have calculated are as follows top middle bott om Mass flux 10 - 4 (kg m - 2 s - 1)

. Analysis of your figure 4.

Data points are taken from your figure 4, lines are exponential fits. Text shows the calculated mass flux. Colours are as follows: red (top of cloud); green (middle); blue (bottom of cloud).

We should expect that the mass flux increases if the particles grow by vapour diffusion, or decreases if the particles evaporate. If vapour diffusion is not important we should observe that the mass flux is conserved. Here we see approximately a factor of 1.7 reduction in the mass flux in the middle of the cloud. I suspect that these numbers are within the expected retrieval errors (or errors in mass - dimension / velocity ‐ dimension relations, but it would b e useful if you could comment on this. The fact that I have used a velocity ‐ dimension power law that is not based on your observation may also be responsible for this too: another reason why it would be helpful to see your velocity - size relations. N ext I thought I would try the analysis of Mitchell (1988) to attempt to calculate the aggregation efficiency. The relevant equation is equation 16 in Mitchell (1988). which can be rearranged for E_a, the aggregation efficiency. Here, β =1.9, b=0.33, a=6.96, α =0.0185 (SI units); λ is the slope of the size - distribution; χ_f is the mass flux (the mass falling through an area per second); Γ is the gamma function and I_1 is a definite integral to be calculated (see Ferrier et al 1994)

From the data in Figure 1 I was able to estimate !" !" to be 7.9 (SI units); λ =6.42e3; and χ_f =2.39e - 4 (SI units) are based on values in the middle of the cloud. I calculated the integral, I_1, as 37.89 – code can be provided on request – feel free to contact me. From these numbers , and rearranging Mitchell's equation above, one can estimate Ea to be equal to approximately 0.4. This number is not too far from what you have used, but it would be useful to understand where the differences arise – I think your estimate is a little higher . For instance on page 12 you say you also use Mitchell (1988); hence, I wondered whether you could go through the calculation in more detail. I suspect this is due to the power laws used for velocity – size , but it may also be due to error s in fitting slope and intercept parameters to the data for instance. I was not sure whether you had taken into account diffusional growth either. Taking into account diffusional growth with increase the slope, so the aggregation efficiency will need to be higher than I have calculated to lead to the observed reduction in the slope. Additionally my estimate of Ea=0.4 assumes the mass flux in the middle of the cloud to be 2.4e - 4, which is low compared to the top and bottom. If I use the higher mass flux 4.8e - 4 (the value I calculated from your data at cloud base) , in the calculation, the corresponding Ea is approximately 0.2. In addition I thought I would try and reproduce a plot similar to your figure 5c. My Figure 2 shows these simulations using aggregation efficiencies of 1, 0.4 and 0.2. The finding here is that lower values of the aggregation efficiency yield lambda values closer to your observations at the 4 km level. Again I think the reason for this discrepancy may be because my calculations have used a terminal fall speed power law that does not match the observations for small crystals. Since the calculations appear to be quite sensitive to the terminal fall speed relation it would be really useful if you could present the measured fall speed (and regression coefficients) you have used.

Figure 2 .

Model simulation using the initial conditions taken from the top of the cloud in figure 4, using different values of the aggregation coefficient.

Final word – I strongly support the statement about sizing particles down to 0.3mm, which would allow you to probe earlier stages of aggregation.

Rapid ice aggregation process revealed through triple-wavelength Doppler spectra radar analysis

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Abstract.

~~Rapid~~ We have identified a region of an ice cloud where a sharp transition of dual-wavelength ratio occurs at a fixed-height for longer than 20 minutes. In this paper we provide evidence that rapid aggregation of ice particles ~~has been identified by combining data from three co-located, vertically-pointing radars operating at different frequencies. A new technique has been developed that uses~~ occurred in this region creating large particles. This evidence comes from triple-wavelength Doppler spectra radar data that were fortuitously being collected. Through quantitative comparison of the Doppler spectra from ~~these radars to retrieve the vertical profile of~~ the three radars we are able to estimate the ice particle size distribution (of particles larger than 0.75 mm) at different heights in the cloud. This allows us to investigate the evolution of the ice particle size ~~distributions.~~ distribution and determine whether the evolution is consistent with aggregation, riming or vapour deposition. The newly-developed method allows us to isolate the signal from the larger (non-Rayleigh scattering) particles in the distribution. Therefore, a particle size distribution retrieval is possible in areas of the cloud where the dual-wavelength ratio method would fail because the bulk dual-wavelength ratio value is too close to zero.

The ice particles grow rapidly from a maximum size of 0.75 mm to 5 mm while falling less than 500 m and in under 10 minutes. This rapid growth is shown to agree well with theoretical estimates of aggregation, with aggregation efficiency ~~close to 1~~ approximately 0.7, and is inconsistent with other growth processes, e.g. growth by ~~deposition,~~ vapour deposition or riming. The aggregation occurs in the middle of the cloud, and is not present throughout the entire lifetime of the cloud. However, the layer of rapid aggregation is very well defined, at a constant height, where the temperature is ~~-15°~~ −15 °C, and lasts for at least 20 minutes (approximate horizontal distance: 24 km). Immediately above this layer, the radar Doppler ~~spectra~~ spectrum is bi-modal, which signals the formation of new small ice particles at that height. We suggest that these newly formed particles, at approximately ~~−15°~~ −15 °C, grow dendritic arms, enabling them to easily interlock and accelerate the aggregation process. The large estimated aggregation efficiency in ~~the studied cloud is between 0.7 and 1,~~ this cloud is consistent with recent laboratory studies for dendrites at this temperature.

~~A newly developed method for retrieving the ice particle size distribution using the Doppler spectra allows these retrievals in a much larger fraction of the cloud than existing DWR methods. Through quantitative comparison of the Doppler spectra from the three radars we are able to estimate the ice particle size distribution at different heights in the cloud. Comparison of these size distributions with those calculated with more basic radar-derived values and more restrictive assumptions agree very~~

well; however, the newly developed method allows size distribution retrieval in a larger fraction of the cloud because it allows us to isolate the signal from the larger (non-Rayleigh scattering) particles in the distribution and allows for deviation from the assumed shape of the distribution.

1 Introduction

Ice microphysical processes are an important part of cloud and precipitation formation; most surface precipitation begins as ice particles (Field and Heymsfield, 2015). However, numerical models, of either weather or climate, have difficulty accurately simulating ice cloud. For example, the CMIP5 models have regional cloud ice water paths that differ from observations by factors of 2–10 (Li et al., 2012). This challenge is partly because observations of ice particles are sparse and because processes controlling the formation and evolution of ice particles, such as aggregation, are poorly understood and crudely parameterized in most models.

Measuring Additionally, measuring the number and size of ice particles within clouds is challenging. The two main methods, in-situ aircraft observations, and active remote sensing observations, both have their deficiencies. First, active remote sensing instruments, such as radar and lidar, are good at measuring the bulk scattering quantities, such as radar reflectivity. However, converting from these bulk quantities to cloud microphysical properties requires numerous assumptions . (e.g. the shape of individual hydrometeors, the particle size distribution). In contrast, aircraft observations measure the size and number of ice particles directly, but only within a small sample volume, at a single height at any given time, and only during sporadic case studies. Furthermore, ice particle size distributions have been shown to be biased as a result of shattering of ice particles on aircraft-mounted instrument inlets (Westbrook and Illingworth, 2009; Korolev et al., 2011), which results in an artificially increased concentration of small ice crystals.

Nevertheless, cloud microphysical observations and in particular particle size distributions are important for many applications. One important application is the better understanding of processes that occur within clouds. For example, size distributions measured from aircraft have been used to study aggregation in cirrus clouds (Field et al., 2006). Furthermore, the size distribution itself affects the relative importance of vapor deposition, riming and aggregation of ice particles. for ice particle growth. Vapor deposition and evaporation rates are proportional to first moment of particle-size distribution, while riming is related to higher moments (product of projected area and fall speed), while aggregation rates depend on the breadth of the particle-size distribution through the difference in fall speeds. So the shape and breadth of the particle-size distribution are an important control on the relative importance of the processes involved. Another important application is to provide observations with which numerical models can be evaluated and their parameterizations improved.

In this paper, we present observations of rapid changes of the ice microphysical properties that report of radar observations of one cloud system, where large vertical gradients in cloud microphysical properties were observed at a fixed height for at least 20 minutes. By exploring the radar data beyond the standard bulk quantities, and exploiting observations from multiple radars together with their Doppler spectra, we are able to estimate the size distribution of particles at different heights and therefore diagnose the most likely process for the rapid but consistent changes in cloud properties with height. The changes of cloud microphysical properties with height apparently result from rapid aggregation of ice particles. These observations were made using three co-located, vertically pointing radars at different frequencies (3, 35, 94 GHz). ~~We are able to retrieve the ice particle size and number concentration through comparison of the Doppler spectra returns from each of the three radars.~~

Analysis of the radar Doppler spectra has previously been performed for the onset of drizzle in stratiform clouds (Kollias et al., 2011a, b) and the application of multi-frequency Doppler spectra has been used to determine the rain size distribution (Tridon and Battaglia, 2015; Tridon et al., 2017). For the ice phase, the three different frequencies have been used simultaneously to categorize rimed and unrimed particles from the surface (Kneifel et al., 2011, 2015, 2016) and from aircraft-based radar observations ~~(Kulie et al., 2014)~~(Kulie et al., 2014; Leinonen et al., 2018; Chase et al., 2018). However, this is the first attempt to retrieve the ice particle size distributions from multi-frequency Doppler spectra observations. These retrievals are then used to evaluate the microphysical processes active within the clouds.

The aggregation process can be characterised by the aggregation kernel k (Mitchell, 1988, eq. 9)

$$k = \frac{\pi}{4} E_{\mathrm{agg}} \left(D_1 + D_2\right)^2 \left|v(D_1) - v(D_2)\right| , \tag{1}$$

where D_1 and D_2 are the diameters of the two potentially-aggregating particles and $v(D)$ is the fall velocity of the particle. The aggregation efficiency of ice particles (E_{agg}; the probability that two ~~colliding particles will~~ particles experiencing a ”close approach´´ will collide and stick together) are typically low, although a large range of values have been reported and understanding of how aggregation efficiency varies with environmental parameters is still sparse. E_{agg} has previously been found to depend on both the particle habit and the temperature at which the collisions occur; however, a large range of values have been reported. ~~A laboratory study (Hosler and Hallgren, 1960)~~ An increase of the aggregation efficiency at about −15 °C has been reported in several laboratory studies. One such study, (Hosler and Hallgren, 1960), where small particles were drawn past a large stationary ice target showed a weak temperature dependence of E_{agg} with a broad peak around ~~−12°~~−12 °C and maximum values of 0.1–0.2. Connolly et al. (2012) used a 10-m tall cloud chamber containing large concentrations of small ice particles settling under gravity and reported a much sharper peak of E_{agg} around ~~−15°~~−15 °C, with values of 0.4–0.9, but ~~values below 0.2 at all other temperatures .~~ the best estimate at other temperatures was below 0.2. Keith and Saunders (1989) found aggregation efficiencies for planar snow crystals drawn past a cylindrical target of 0.3–0.85 depending on the particle size. Hobbs et al. (1974) reported that both the maximum dimension of ice aggregates and the probability of seeing aggregates increased at around ~~−15°~~−15 °C, which was linked to the preferred formation of dendritic particles at this temperature. This is supported by other studies showing larger E_{agg} in the presence of dendritic particles. Mitchell et al. (2006) found E_{agg} of around 0.55 for clouds dominated by dendrites at cloud top, but much lower values around 0.07 when dendrites were not present. Low E_{agg} values of 0.09 were also found for tropical anvil clouds where dendritic particles were not present at temperatures of ~~−3°C to −11°~~−3 °C to −11 °C (Field et al., 2006). In the early stage of aggregation, Moisseev et al. (2015) reported that the aggregates were made up of a small number of dendritic particles. These studies seem to suggest that dendrites, which typically form at around ~~−15°~~−15 °C, can significantly increase the aggregation efficiency because the dendritic branches interlock with other particles, whereas the aggregation efficiency is much lower when dendritic particles are not present. In this study, retrievals from radar observations will be used to estimate the aggregation efficiency and will be compared with the laboratory-derived values.

Barrett et al. (2017) showed that the assumed particle size distribution is the single-largest sensitivity in the model physics for mixed-phase altocumulus clouds. The importance of correctly simulating the ice particle size distribution has been shown in several other studies (Pinto, 1998; Harrington et al., 1999; Field et al., 2005; Morrison and Pinto, 2006; Solomon et al., 2009). Therefore understanding and correctly implementing the aggregation process in numerical models of cloud physics is important for the overall development of the cloud system.

This paper is organised with an overview of the instruments and data in section 2, an overview of the case study in section 3 and details about the retrieval in section 4. Section 5 details the cloud properties retrieved and their uncertaities and section 6 summarizes the evidence for aggregation, with conclusions drawn in section 7.

2 Data and Methods

We use data from three co-located radars at the Chilbolton Observatory in Hampshire, Southern England ~~.~~ on the afternoon of 17 April 2014. The radars operate at frequencies of 3 GHz ~~(9.75-cm wavelength, 25-m antenna, 0.28° beamwidth),~~ (9.75-cm wavelength, 25-m , 35 GHz ~~(8.58-mm wavelength, 2.4-m antenna, 0.25° beamwidth)~~ (8.58-mm wavelength, 2.4-m antenna, 0.25° beamwidth; Illingworth et a and 94 GHz ~~(3.19-mm wavelength, 0.46-m antenna, 0.5° beamwidth)~~(3.19-mm wavelength, 0.46-m antenna, 0.5° beamwidth; Eastment, 1 . The 35- and 94-GHz cloud radars are situated immediately next to one another, whereas the 3-GHz radar is sited less than 50 m away (Fig. 2). The sampling of the three radars was synchronized to within 0.1 seconds and full pulse-to-pulse power and phase measurements were recorded. For the 3-GHz radar, Doppler spectra were calculated every second and incoherently averaged over 10 seconds. For the 35-GHz and 94-GHz cloud radars, spectra were calculated every 0.11 and 0.08 seconds respectively and again incoherently averaged over 10 seconds. Assuming typical wind speeds of 20 ~~ms-1~~ m s^{-1} aloft, the averaged spectra correspond to a 200-m section of cloud. Ground clutter was removed from the spectra by masking returns with velocity near zero. Noise levels were estimated from measurements beyond the range of meteorological echoes (> 10 km) and subtracted from the individual spectra prior to averaging. The data from each radar was interpolated ~~onto~~ on to common range and velocity grids (60-m range by 0.0195 m s^{-1} velocity).

Because of the large antenna, it is necessary to apply a near-field correction to the 3-GHz data at heights below about 6~~km~~ km (Sekelsky, 2002). This correction factor was derived empirically by comparing 3-GHz reflectivity profiles against those measured by the 35-GHz instrument (which has a much smaller antenna) in a number of Rayleigh scattering ice clouds. The magnitude of the correction was 1 dB at 5 km, rising to 3 dB at 3 km.

2.1 Data quality, calibration and attenuation correction

To account for potentially imperfect calibration and attenuation by atmospheric gases and liquid water in the lower troposphere, the 35- and 94-GHz reflectivity is corrected relative to 3-GHz radar. The 3-GHz radar is absolutely calibrated to within 0.5 dB, using the method of Goddard et al. (1994b). The radar reflectivity value from the cloud radars (35 and 94 GHz) was adjusted to

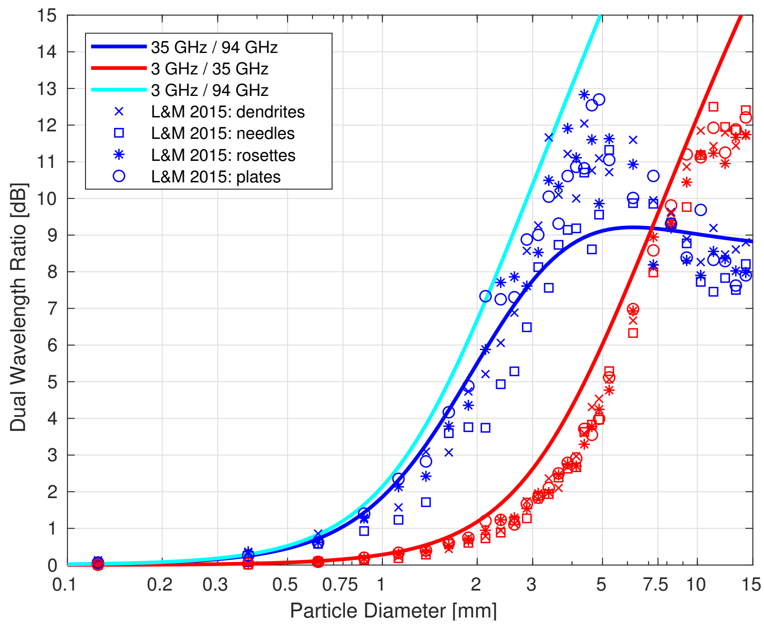

Figure 1. Dual-wavelength ratios as a function of ice particle diameter for the three pairs of radar frequencies used in this study. Dual-wavelength ratios from the Westbrook et al. (2006, 2008a) scattering model are shown with solid lines. For comparison, mean dual-wavelength ratios of unrimed aggregates within 250-micron-wide diameter bins from Leinonen and Moisseev (2015) are shown by points for four different aggregate-monomer types.

Table 1. A summary of the terminology used throughout this paper, where F denotes the radar frequency.

symbol	variable name	variable definition	~~units~~ unit
Z_F	radar reflectivity	total radar cross sectional area of scatterers within the target volume	dBZ [Z = ~~mm^{6}m^{-3}~~mm^6 m^{-3}]
$DWR_{F1/F2}$	dual-wavelength ratio	$Z_{F1} - Z_{F2}$	dB
sZ_F	spectral reflectivity	radar reflectivity per Doppler-spectra velocity bin	dBX [X = ~~mm^{6}m^{-3}(m s^{-1})$^{-1}$~~mm^6 m^{-3} (m s^{-1})$^{-1}$]
$sDWR_{F1/F2}$	spectral dual-wavelength ratio	$sZ_{F1} - sZ_{F2}$	dB

match the 3-GHz radar reflectivity in each profile so as to remove any calibration or attenuation offsets. The adjustment amount was estimated in regions where Rayleigh scattering was expected at all three wavelengths[1] and hence where the reflectivity should be the same from each radar. The adjustments ~~made~~ reduce the median difference in reflectivity (Z) in the Rayleigh scattering areas to 0 dB. The same adjustment to Z (in dB) is made throughout the profile. A different correction is applied individually to each 10-second profile; the equivalent dB correction is also applied to the ~~spectra within the profile.~~ Doppler spectra power within each profile. This adjustment works well because the majority of the attenuation by atmospheric gases and liquid water occurs below cloud base. In other cases, where cloud base is lower or with embedded liquid water layers, a different treatment would be necessary.

The multi-wavelength approach allows us to measure the diameter of ice particles that are comparable in size to the shortest radar wavelength, or larger (e.g. Kneifel et al., 2015, 2016). For ice particles comparable in size to the radar wavelength, non-Rayleigh scattering becomes important. For suitably large particles, it becomes possible to size the particles based on the different radar returns at different wavelengths.

~~The~~ In contrast to the bulk retrieval that makes a single retrieval for particles of all fall velocities together, the Doppler spectra approach allows for retrievals of particle size and number concentration to be made separately on particles of distinct fall velocities. We can use the multi-wavelength approach to determine the representative particle size from the "spectral dual-wavelength ratio" (sDWR; i.e. the difference in reflectivity of particles within a small range of fall velocities; see Table 1 for a full summary of radar quantities used in the paper), but can additionally separate the particles based on their fall velocity allowing us to retrieve the ice particle size distribution.

A correction to the velocities measured by the radar is also applied. Unfortunately, the three radars were not precisely vertically pointing for this case (as determined by biases in the mean Doppler velocity in the Rayleigh-scattering part of the cloud) and initial testing suggested that there was a large sensitivity to the velocity offsets in the spectra (see section 6.1). The 3-GHz radar was pointing vertically, but after analyzing the data, the 35- and 94-GHz radars were determined to be off-zenith by approximately 0.2° and 0.15° respectively in opposing directions. ~~This mis-pointing~~ These offsets were determined by assessing the mean Doppler-velocity differences between the three radars as a function of height. The correlation of these velocity differences with the atmospheric wind profile (determined from ECMWF forecast fields) enabled an estimation of the pointing angle errors.

The mispointing of the radars is small and likely does not result in a substantial ~~mis-match~~ mismatch in sample volume given the 10-second integration time. However, this small ~~mis-pointing~~ mispointing means that the radar detects a small component of the horizontal wind in addition to the fall velocities of the ice particles. Although the pointing angle error is small, the horizontal wind component detected is of the order of a few centimeters per second, which is sufficiently large to affect our comparison of the Doppler spectra from the three radars. Therefore, we have made a correction to the velocity measurements for the 35- and 94-GHz radars to ensure that the spectra are well aligned and can be compared. This correction is important because even a small shift in velocity can substantially affect the estimates of sDWR. In practice, the correction applied is $+0.0585$ m s^{-1} (3 velocity bins) for the 35-GHz radar and -0.0390 m s^{-1} (2 velocity bins) for the 94-GHz radar throughout the cloud layer. This correction is imperfect; however, we do not have independent measurements of the horizontal wind speed with sufficient accuracy and high enough vertical resolution to make a reliable height-dependent correction or indeed any direct measurement of the mispointing. Radiosonde data and ECMWF model output shows that the horizontal wind speed was

[1] Based on an analysis of reflectivity differences, Rayleigh scattering is assumed where the 3-GHz reflectivity is below 5 dBZ and the absolute difference between the 3- and 94-GHz velocity measurements is less than 0.025 m s^{-1}. Measurements were also excluded where the 3-GHz reflectivity was less than -10 dBZ to avoid effects of residual ground clutter.

near-constant with height throughout the cloud layer on this day, and inspection of many individual Doppler spectra indicate that our simple correction aligns the spectra very well in this case~~(see Figure 5a,d,g)~~.

To reduce the noise in the spectra, each individual spectrum has been smoothed in velocity space by averaging over a 0.0585 m s^{-1} window, which equates to three velocity bins.

We mask out regions where significant turbulence is present because the vertical air motions are large and vary on small time and space scales compared to the particle fall velocities that we are trying to measure. Near the cloud base, there is a layer of substantial turbulence caused by ~~ice particles subliming~~ sublimation of ice particles as they fall into ~~drier air and leading~~ subsaturated air and this leads to destabilisation of the atmosphere in this layer. In this turbulent layer, the implicit assumption that measurements at a specific velocity are of a single particle size is invalid~~and Rayleigh scattering is assumed where the 3-GHz reflectivity is between −10 and +5 dBZ~~. Hence, we identify regions where turbulence is altering the spectra, ~~based on~~ by calculating the contribution of turbulence to spectral width using O'Connor et al. (2005, eqns. 10–15). Points where the velocity variance from turbulence exceed a threshold value of 10^{-3} m^2 s^{-2} are not considered when performing our retrievals. This threshold value was chosen such that all affected regions were suitably masked and in the remaining data that the width of the Doppler spectrum was determined by microphysical rather than turbulent contributions. Additionally, any points in the spectra that are 20 dB down from the peak of the spectra are removed in order to minimise the impact of noise.

3 The case - 17 April 2014

Figure 3a shows the radar reflectivity measured at Chilbolton for the thick stratiform ice cloud observed on 17 April 2014. This cloud formed in north-westerly flow, ahead of a cold front. The surface cold front reached Chilbolton at about 1800 UTC. The front was not associated with any surface precipitation at Chilbolton, and only very light precipitation across some other parts of Southern England.

The ~~cloud~~ evolution of the cloud reflectivity and the ratio of 35-GHz and 94-GHz reflectivity are shown in Figure 3. The cloud top height was approximately 9 km, where 35-GHz reflectivity values are around −15 dBZ, and increase to 19 dB at approximately 4 km altitude, near cloud base. The temperature at cloud top was ~~−45°~~ −45 °C and the freezing level was at about 2.7 km.

~~The evolution~~ Throughout most of the cloud ~~reflectivity and the ratio of 35-GHz and 94-GHz reflectivity are shown in Figure 3. The~~ the DWR values are below 1 dB. However, at around 4 km altitude there is a rapid increase of DWR with decreasing height which indicates an increase in particle size such that the backscattered return at 94 GHz is no longer from Rayleigh scattering. The region of these large DWR values is consistent in height (onset at 4.5 km altitude; Fig. 3c), and is evident for at least 35 minutes. The largest DWR values occur at around 1615 UTC, with peak values reaching 7 dB. The profile of DWR values at 1615 UTC is shown in Fig. 3c.

Radar data from a larger portion of the same cloud was analysed in Stein et al. (2015)~~, who~~ . Earlier in the day (before 1540 UTC), the cloud did not show this sharp transition to high DWR values around 4.5 km. Stein et al. (2015) also used the triple-frequency radar data to determine that the cloud contained primarily aggregate snowflakes, consistent with the Westbrook et al.

Figure 2. A photograph of the three co-located radars at the Chilbolton Observatory, Hampshire, England. From left to right: the 3-GHz CAMRa radar, 94-GHz radar and 35-GHz radar.

(2006, 2008a) scattering model ~~.~~ (lines in Fig. 1). Scattering properties of unrimed aggregates from Leinonen and Moisseev (2015) are also consistent with observations, and give very similar characteristics to the Westbrook et al. (2006, 2008a) scattering model (points in Fig. 1). We focus on the ~~the~~ time from 1545 to 1620 UTC, where there are dual wavelength ratios up to 8 dB below 4.5 km (Fig. 3b~~).~~ ,c).

We attempt to understand what causes the rapid change in cloud properties during this period of substantial $DWR_{35/94}$ and the rapid change in height. Looking at the spectral reflectivity at each height (sZ_{35}; Fig. 4a) together with the spectral dual wavelength ratio ($sDWR_{35/94}$; Fig. 4b) reveals the changes of the cloud properties with altitude. From these data, the origin of the large changes in the sharp transition can be identified. At 5.4 km, there is an increase in the signal coming from slow-falling

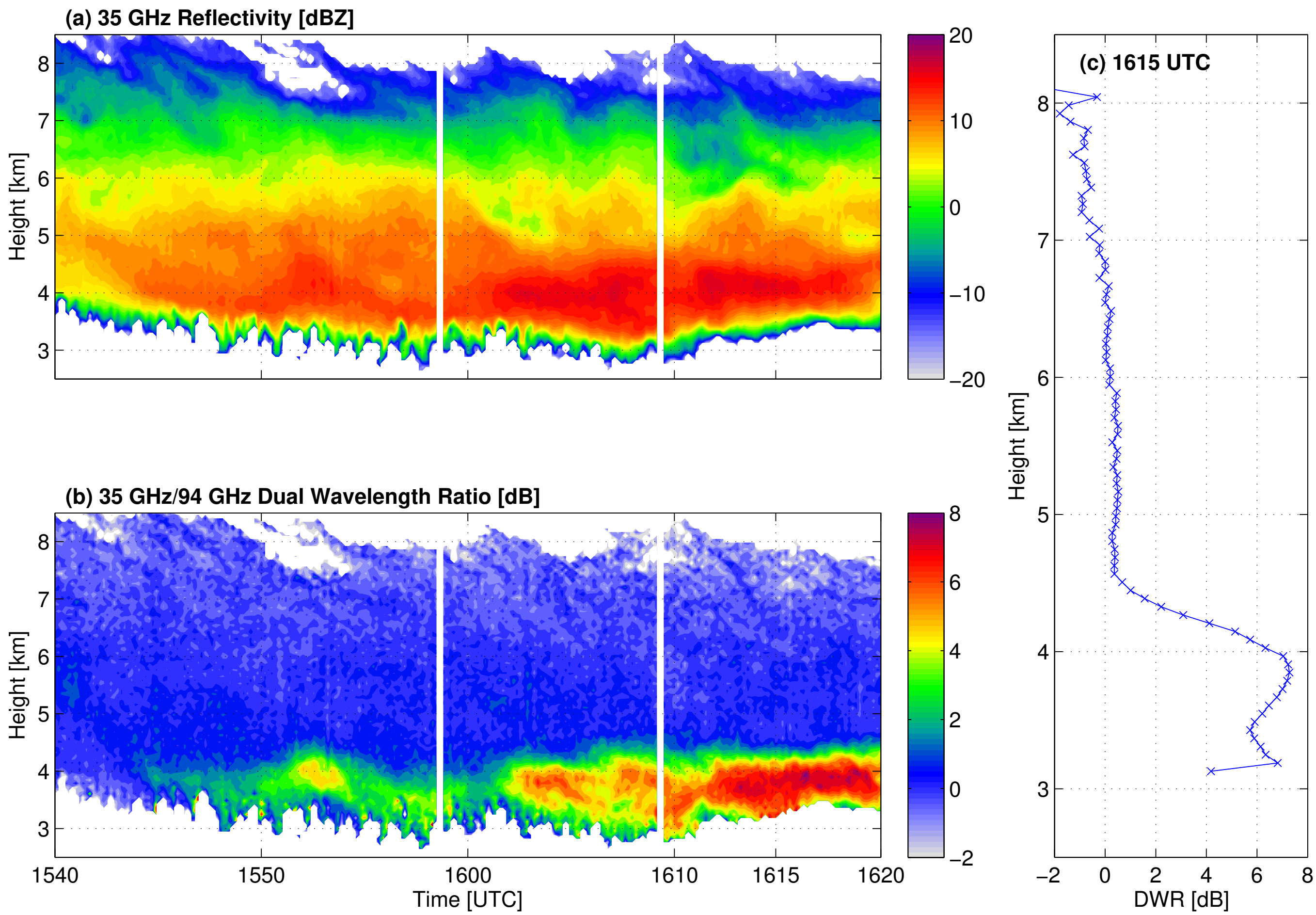

Figure 3. Overview of the cloud structure on 17 April 2014 showing the a) 35-GHz radar reflectivity and b) the ratio of 35-GHz reflectivity to 94-GHz reflectivity throughout the sampling period. c) The vertical profile of DWR at 1615 UTC.

particles (0.4–0.6 ms^{-1}; Fig. 4a). At this height, only the fastest falling particles have $sDWR_{35/94} > 1dB$. At 4.5 km, the reflectivity and spectral reflectivity of the slow-falling particles has increased. The $sDWR_{35/94}$ increases up to 8 dB for the fastest-falling particles, and by 4 km the increase in $sDWR_{35/94}$ is seen for the majority of particles. Interestingly, the fall velocity of these particles does not increase as the particles grow larger and produce large $sDWR_{35/94}$ values.

4 ~~Retrieving~~ Retrieval of the ice particle size distribution

To retrieve the ice particle size distribution from the ~~Doppler spectra at three wavelength~~cross-calibrated and velocity-matched Doppler spectra (see section 2.1) at three wavelengths, we use the method described below. The method is illustrated at three separate heights in Figure 5. At a given range gate weThe following is calculated for each individual velocity bin, within each radar range gate and at all times:

1. Calculate the spectral dual-wavelength ratio (sDWR $= sZ_{35} - sZ_{94}$)between . This is simply calculated as the difference between the spectral reflectivity (sZ) at 35 and 94 GHz (Fig. 5a,d,g).

2. Use a scattering model Determine the particle diameter D from sDWR. The relationship between particle diameter and particle DWR from the Westbrook et al. (2006, 2008a) scattering model (Fig. 1) is used to convert the sDWR value to a particle sizeparticle diameter. We use the Westbrook et al. (2006, 2008a) scattering model , this scattering model based on its good agreement with observational data for this case (Stein et al., 2015). Additionally, the scattering model is used to calculate the reflectivity value for single particles of the retrieved size. (We assume the ; other scattering models may be more appropriate for different cases.

3. Calculate the mass m of an ice particle with diameter D, assuming the Brown and Francis (1995) mass-size relationship of $m = 0.0185D^{1.9}$ for all ice particles).

4. Use the single-particle reflectivity calculated in the previous step and the total reflectivity measured by the radar to calculate the . Use of this mass-diameter relationship is supported by (Stein et al., 2015), who found that the fractal dimension of snowflakes on this day was 1.9 and hence the exponent of 1.9 is appropriate; other mass-diameter relationships may be more appropriate for different cases.

5. Determine the radar reflectivity of a single ice particle with diameter D and mass m using the scattering model.

6. Determine the total number of particles of each size. within the velocity bin. This is calculated by dividing the total spectral reflectivity sZ by the single-particle reflectivity calculated in the previous step.

The size and number of ice particles within each the velocity bin is now known(Fig. 5c,f,i). The particle size distribution can be estimated by performing this same process for each velocity bin.

Using this method Up to this point, we have determined the diameter D of the ice particles D within each velocity bin, and also the particle velocity size distribution dN/dV (where dN is the concentration of ice particles with velocity between V and V+dV). We can convert dN/dV to the ice particle size distribution dN/dD (concentration of ice particles in a diameter bin, normalized by the bin widthwith diameter between D and D+dD). This is the common way to express a particle size distribution that is independent of the measuring sample interval (dD or dV). To do so, we need to know the relationship between the velocity bin width dV and the diameter bin width dD. To determine this, we use a 300-m by 90-s window (5 range gates by 9 individual averaged spectra) centered on the current radar pixel and compute the optimal power-law fit to the velocity and measured Doppler velocity and retrieved diameter values, of the form $V = cD^d$. A power-law relationship is used because it is both easily differentiable and common in microphysical scaling relationships (e.g. Locatelli and Hobbs, 1974). We use the differential of this power-law fit to compute $\frac{dV}{dD}$ - the diameter bin width for each velocity bin. The size distribution is then calculated as

$$\frac{\mathrm{d}N}{\mathrm{d}D} = \frac{\mathrm{d}N}{\mathrm{d}V}\frac{\mathrm{d}V}{\mathrm{d}D}. \tag{2}$$

There is a relatively large sensitivity of the retrieved size distribution to the power-law fit, but ~~only~~ this is primarily in terms of the ~~absolute~~ number concentration, rather than the diameter of the particles or the shape of the size distribution (see section 6.1 for a complete sensitivity analysis).

~~The retrieved~~ Retrieval of the size and number concentration of ice particles is only possible for particles larger than about 0.75 mm in diameter (corresponding to a $\mathrm{sDWR}_{35/94}$ of about 1 dB, Fig. 1). For smaller particles~~the radar returns~~, sZ is very similar at all three ~~frequencies are small and~~ radar frequencies and differences are not easily distinguished from noise in the spectra. For particles larger than about 3 mm diameter, the $\mathrm{sDWR}_{35/94}$ saturates at about 8–9 dB (Fig. 1) as a result of the fractal geometry of the aggregates (see Stein et al., 2015) and therefore retrieval of particle diameter from $s\mathrm{DWR} - 35/94$ is no longer possible. Therefore, where $\mathrm{sDWR}_{35/94}$ is larger than 6 dB, the diameter and number concentration are retrieved using sDWR~~$_{3/94}$ using~~$_{3/35}$ instead, following the same method as above. This pair of frequencies does not saturate until significantly larger particle diameters and therefore, for larger particles, has a larger sensitivity to change in diameter than for the 35/94 GHz pair. We do not use the 3/35 GHz pair for the full range of particle diameters because the 3-GHz is affected more by noise than the 35-GHz spectra, and therefore negatively impacts the retrieval of particle sizes when DWR is small. It would be equally valid to calculate the size and number concentration of the larger particles using the 3/94-GHz pair instead, and this indeed enables a consistency check that the retrieval works well and that the input Doppler spectra are well matched.

5 Retrieved cloud properties and validation

Throughout most of the cloud, the 35/94-GHz dual-wavelength ratio ($\mathrm{DWR}_{35/94}$) is near zero (<1 dB) (Fig 3b), implying that the ice particles are relatively small and are still in the Rayleigh scattering regime at 94 GHz (~~max~~ maximum diameter 0.75 mm). DWR only exceeds 2 dB after 1545 UTC and between 4.3 km and cloud base.

From 1600 to 1620 UTC, there is a sharp transition from $\mathrm{DWR}_{35/94} < 1$ dB at 4.5 km to peak $\mathrm{DWR}_{35/94}$ values at 4 km, with the maximum $\mathrm{DWR}_{35/94} = 8$ dB. The altitude of this sharp transition is consistent after 1602 UTC, with the largest $\mathrm{DWR}_{35/94}$ values being present after 1610 UTC. There is also evidence of this transition layer as early as 1545 UTC.

~~We focus on this period of substantial $\mathrm{DWR}_{35/94}$ and the rapid change in height to investigate the retrieved properties of the clouds and attempt to understand what causes the rapid change in cloud properties. Looking at the spectral reflectivity at each height (Fig. 4a) together with the spectral dual wavelength ratio (Fig. 4b) reveals the changes of the cloud properties with altitude. From these data, the origin of the large changes in the sharp transition can be identified. At 5.4 km, there is an increase in the signal coming from slow-falling particles (0.4–0.6 ms^{-1}; Fig. 4a). At this height, only the fastest falling particles have $\mathrm{sDWR}_{35/94} > 1$dB. At 4.5 km, the reflectivity and spectral reflectivity of the slow-falling particles has increased. The $\mathrm{sDWR}_{35/94}$ increases up to 8 dB for the fastest-falling particles, and by 4 km the increase in $\mathrm{sDWR}_{35/94}$ is seen for the majority~~

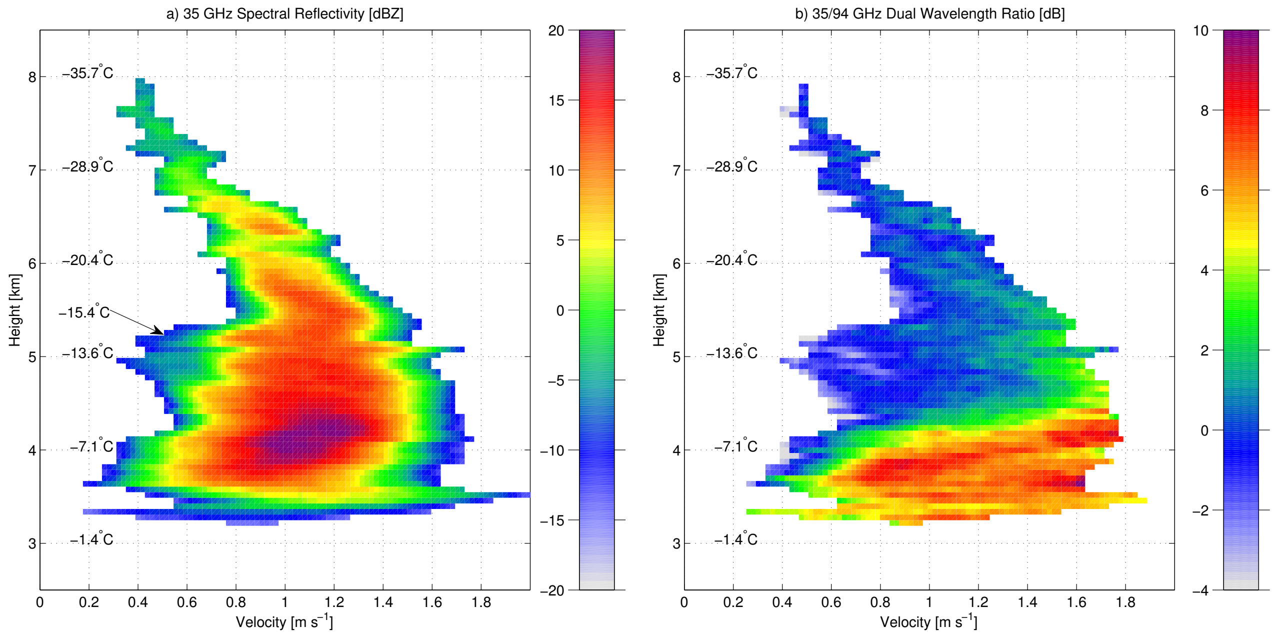

Figure 4. Height profile of a) spectral reflectivity at 35 GHz (sZ_{35}) and b) spectral dual wavelength ratio (sDWR$_{35/94}$) recorded at 1615 UTC. Temperatures from the ECMWF model at 1600 UTC are shown every 1 km and at 5.3 km where the small-particle mode is first evident.

~~of particles. Interestingly, the fall velocity of these particles does not increase as the particles grow larger and produce large sDWR$_{35/94}$ values.~~

More detail can be seen by examining the Doppler spectra for the different radars at a few fixed heights in detail. The Doppler spectra measured at 5.89, 4.81 and 4.15 km (Fig. 5a,d,g) show three spectra with rather different shapes.

At 4.15 km, the spectra has only a single mode but throughout most of the velocity range sZ_{35} is much greater than sZ_{94}. The sDWR$_{35/94}$ reaches 8 dB (Fig. 5h) and the largest particles are sized at around 5 mm. The retrieved size distribution is approximately inverse exponential (Fig. 5i).

At 5.89 km (top row of Fig. 5), in contrast, the spectra for ~~35 and 94 GHz~~ all three radars are very similar with a single peak~~and~~; all sDWR$_{35/94}$ values below 1 dB ~~.~~ (Fig. 5b). The small sDWR$_{35/94}$ values mean that it is not possible to reliably size the ice particles here, other than to say that they are all smaller than 0.75 mm.

About 1 km lower in the cloud, at 4.81 km (second row of Fig. 5), the mean velocity and reflectivity have both increased, but there is also a bi-modal structure to the spectra captured at both frequencies. This second mode is related to newly formed, small ice particles that are falling slower than the majority of older, larger ice particles. Furthermore, at 4.81 km, there are larger and faster-falling particles present than at 5.89 km. The largest sDWR$_{35/94}$ values now approach 4 dB (Fig. 5e), and particles larger than 0.75 mm are present, with the largest retrieved diameter of 1.2 mm. The size distribution (Fig. 5f) of the reliably-sized particles (those larger than 0.75 mm and outside the gray region of the plot) is inverse exponential.

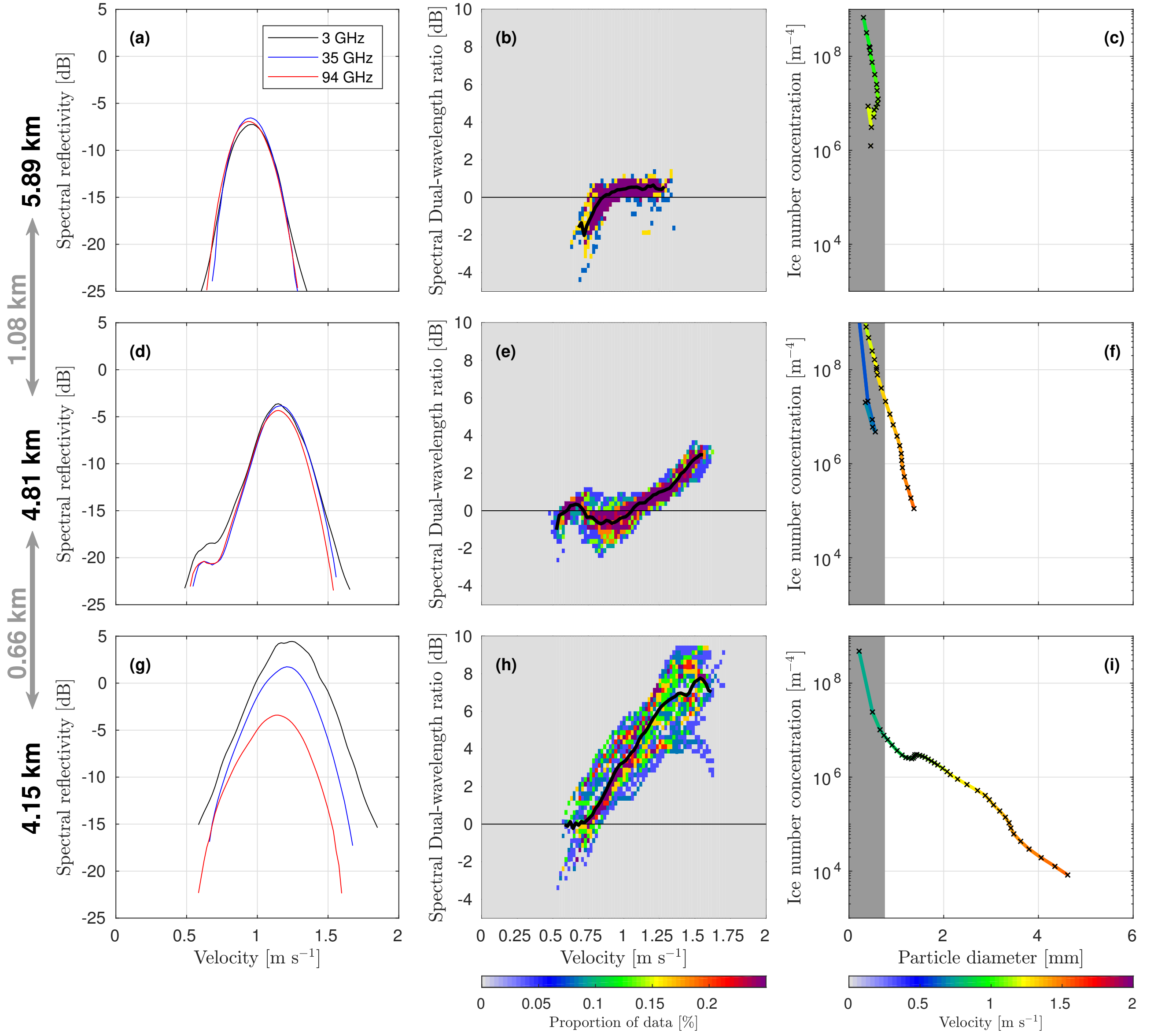

Figure 5. Illustration of the retrieval method and the retrieved size distribution at 3 three heights at 1615 UTC. a-c are just above the layer of secondary ice nucleation, d-f are within that layer and g-i are below this layer, where the dual-wavelength differences are largest. a,d,g show the 3-, 35- and 94-GHz spectra at that height. b,e,h show the distribution of $sDWR_{35/94}$ data points within a window around the central time (90-s by 300-m), with the black line denoting the median power difference for each velocity bin. c,f,i show the retrieved ice particle size distribution, with the color of the line related relates to the velocity of the data used to determine that data point. The gray shaded region marks particle diameters smaller than 0.75 mm, where there is no reliable information available to size the ice particles. The higher altitude plots are from earlier times to account for an approximately 1 m s^{-1} fall velocity of the ice particles.

The consistent and narrow range of heights over which this rapid change in size occurs is just below the region where new particles are seen around 5.4 km and the Doppler spectra is bi-modal (Fig. 5~~a~~d). These new particles fall slowly, which suggests that they are small and are ~~nucleated~~ formed at this level. These particles begin to fall faster as they grow in size. Particles forming around ~~−15°~~ −15 °C would initially grow as dendrites (Takahashi et al., 1991). As these particles grow, the $\mathrm{sDWR}_{35/94}$ starts to increase for the larger (faster falling) particles, which we take to be aggregates. This increase in $\mathrm{sDWR}_{35/94}$ implies an acceleration of the aggregation process at this height.

The reduction of the size distribution slope between 4.81 km and 4.15 km remains consistent for at least 30 minutes from 1545 UTC onwards, but is not present earlier in the cloud. The observations shown in Fig. 5 are similar throughout this time period, which explains the sharp increase of DWR between 4.8 and 4.1 km (Fig. 3) during this time period.

5.1 Evolution and validation of retrieved size distributions

To evaluate how accurate the retrieved ice particle size distributions are, we would ideally like to compare against in-situ data. However, in-situ observations are not available for this case. Therefore, we evaluate the retrievals against other retrieval methods.

By fitting an inverse-exponential to the retrieved particle size distribution data from our Doppler spectra method, we can estimate the slope of the size distribution, Λ in $\mathrm{d}N/\mathrm{d}D = N_0 \exp(-\Lambda D)$ (Fig. 6a). By means of verification, we also calculate the ~~the~~ slope of a purely inverse-exponential size distribution fitted to match the $\mathrm{DWR}_{35/94}$ values only (Fig. 6b). There is excellent agreement between the two methods in the regions where the size distribution is broader and less steep. Fig. 6c shows a ~~3-minute~~ 2-minute average of Λ, which again shows the excellent agreement throughout the whole profile, particularly the height of the rapid change of Λ between 5 and 4 km. The only region of disagreement is just below 4 km, where the spectra method suggests even broader size distribution than the DWR method. This could be evidence that the inverse-exponential size distribution approximation in this region is not appropriate or because $\mathrm{DWR}_3 5/94$ has almost saturated at 8–9 dB. However, both methods agree that there is a rapid increase in ice particle size occurring as they fall from 4.5 km to 3.6 km and a broadening of the ice particle size distribution. In the next section, we present evidence that this rapid change is occurring as a result of aggregation and not occurring through vapor deposition or riming.

The spectral method developed here is more sensitive to the presence of a few large particles than the DWR method. With the spectral method, the influence of a few non-Rayleigh scatterers can be seen in the spectra before the reflectivity of the individual scatterers is large enough to contribute significantly to the total reflectivity (which is a weighted average of sDWR over all particles). Therefore, the retrieved particle size distributions higher in the cloud are more reliable with the spectral method than the DWR method, because we are able to isolate the signal from the larger particles in the distribution. However, the spectral method is sensitive to noise in the spectra, and hence when the overall signal becomes weak, and the noise is therefore a more significant contributor, the retrieved particle size distributions are also noisy.

6 Evidence for rapid aggregation of dendrites

In this section we examine whether the changes in particle size and size distribution could be explained by processes other than aggregation. Specifically we address whether vapor deposition or riming could lead to the observed changes.

Ice particles grow from smaller than 0.75 mm in diameter (DWR<1 dB), above this transition layer, to larger than 5 mm by the time they reach 4 km (Figure 3c). Mean radar Doppler velocities just above this transition layer are 1–1.2 m s^{-1} (Figure 5d), indicating that on average ice particles will take 400–500 seconds to fall from 4.5 to 4 km, although the largest particles responsible for the high large DWR values will fall faster than the average particle.

The growth of ice particles by vapor deposition cannot produce large ice particles sufficiently quickly to match our observations. Calculations using the vapor deposition growth equation from Pruppacher and Klett (1978) are presented to demonstrate this. The equations used were,

$$\frac{\mathrm{d}m}{\mathrm{d}t} = \frac{4\pi\, C\, SS_i\, F}{\left(\frac{L_s}{R_v T} - 1\right)\frac{L_s}{KT} + \frac{R_v T}{e_{si}(T) D}} \quad , \tag{3}$$

$$m = 0.0185 D^{1.9} \quad , \tag{4}$$

where the rate of change of particle mass m with time t is a function of the ice particle capacitance C (assumed $= D/4$ here, following Westbrook et al. (2008b), where D is the diameter), supersaturation with respect to ice SS_i and ventilation coefficient F. $F = 0.65 + 0.44 * 0.6^{0}.33\mathrm{Re}^{0}.5$. Re is the Reynolds number; $\mathrm{Re} = \rho D V(D)/\mu$, calculated from the air density ρ, particle diameter D, terminal velocity $V(D)$ and dynamic viscosity of air μ. Terms on the denominator are the latent heat of sublimation L_s, the specific gas constant for vapor R_v, temperature T, thermal conductivity of air K, and saturated vapor pressure over ice e_{si}, (4) is the Brown and Francis (1995) mass-size relationship.

These calculations, for a liquid-saturated atmosphere at −10° −10 °C, show that typical ice particles (Brown and Francis, 1995) would, at their absolute fastest, take over 40 minutes (2534 seconds) to grow from 0.75 mm to 5 mm in diameter. Similarly, Fukuta and Takahashi (1999) calculate that it takes over 30 minutes to grow a particle of 3 mm through vapor deposition. We therefore can rule out pure vapor deposition as the source of the largest particles, which develop in less than 10 minutes.

Riming of the ice particles by collecting liquid water is another possible explanation; however, there is no evidence of significant supercooled liquid water present at this height. There were no strong backscatter returns in the lidar measurements (not shown) which would indicate the presence of liquid droplets, and the liquid water path measured by the microwave radiometer is below the noise level of the instrument (about 20 g m^{-2}) throughout the observation period. Furthermore, the triple-frequency analysis for the scattering models in Stein et al. (2015) do not show agreement with the expected triple-wavelength signature of rimed particles (Kneifel et al., 2016), but rather for aggregate snow crystals.

The sharp and consistent transition of cloud properties with height after 1545 UTC is therefore likely a result of aggregation. The first indication that aggregation is the most important process in this part of the cloud is the continual decrease of Λ (the slope of the ice particle size distribution) with height down from the top of the transition layer. This change with height indicates that there are more large particles and fewer small particles as the particle size distribution evolves, consistent with aggregation. ~~In the case of vapor deposition or riming, one would expect the size of all particles to increase but the relative number of small and large particles would be mostly unchanged. Following Mitchell (1988), we calculate the~~

$$\frac{d\Lambda}{dz} = \frac{\Lambda}{b\chi_f}\frac{d\chi_f}{dz}\left[1-\frac{2\Gamma(b+\delta+1)\Gamma(b+d+1)}{\Gamma(\delta+1)\Gamma(2b+d+1)}\right] - \frac{\pi E_{\mathrm{agg}} I_l \chi_f \Lambda^{b+d-1}}{4abc\Gamma(b+d+1)\Gamma(2b+d+1)} \tag{5}$$

We calculated the expected change of Λ with height using (5), following Mitchell (1988), for several different values of aggregation efficiency (~~E_{agg}).~~ E_{agg}). In (5), a, b are constants in the mass-diameter relationship $m = aD^b$; c, d are constants from the fall velocity-diameter relationship $V = cD^d$; $\delta = 1.0$ following Mitchell (1988); Γ is the gamma function; χ_f is the snow-flux in kg m^{-2} s^{-1} and I_l is calculated from eqn. 20 of Mitchell (1988), dependent on b and d - in our calculations it takes the value of 11.524. These calculations assume that aggregation ~~is the only process~~ and vapor deposition together are the primary processes affecting the evolution of the size distribution~~and that the ice-mass flux("snowfall rate") is constant with height.~~, and that changes to the total mass are only due to vapor deposition or sublimation not the accretion of liquid drops.

To estimate the aggregation efficiency in this part of the cloud, we need to know the slope of the particle size distribution at the top of the layer and the vertical profile of snow flux. The Λ value is estimated from the retrieved size distribution. The snow flux profile is estimated from the retrieved particle diameters, converted to a mass, multiplied by the measured Doppler velocity and then integrated across the spectra. Using the retrieved profile of size distribution properties ~~at 4.5 km height~~ and snow flux profile at 1615 UTC as input, the expected change ~~with height~~ of Λ with height for E_{agg} values of 0.2, 0.7 and 1.0 are shown in Fig. 6c. The evolution of Λ between 4.5 and 4.0 km altitude, as calculated by either the Doppler spectra method or the simpler DWR method, ~~both fall within the $E_{\mathrm{agg}} = 0.7$ and 1.0 curves. The value of 0.2~~are both consistent with theoretical evolution with E_{agg} around 0.7. The $E_{\mathrm{agg}} = 0.2$, reported in Hosler and Hallgren (1960) cannot reproduce the observed broadening of the size distribution through this shallow layer of cloud, and leads to Λ being overestimated by almost an order of magnitude at 3.5 km. ~~This value of E_{agg}~~ $E_{\mathrm{agg}} = 0.7$ is at the higher end of values reported in the literature. However, Connolly et al. (2012) found $0.4 < E_{\mathrm{agg}} < 0.9$ at ~~$-15°$~~ -15 °C, whereas for all other temperatures sampled the best estimate was $E_{\mathrm{agg}} \leq 0.2$. Similarly, Field et al. (2006) reported that E_{agg} values greater than unity were required for small particles for a good fit to observed aggregation within tropical cirrus anvils. Our results are consistent with the high values of E_{agg} of Connolly et al. (2012) at ~~$-15°$~~ -15 °C, but do not support the $E_{\mathrm{agg}} < 0.2$ reported by Hosler and Hallgren (1960). This suggests that the free-fall experiments in the 10-m cloud chamber ~~are~~ may be more representative of the natural aggregation in the atmosphere than the stationary target experiments of Hosler and Hallgren (1960). Connolly et al. (2012) speculate that the higher E_{agg} at ~~$-15°$~~ -15 °C is because the dendritic branches of the crystals are able to interlock and that this can increase E_{agg} by at least a factor of 3. Increased aggregation efficiency in the presence of dendritic crystals also agrees with observations by Hobbs et al. (1974). Our observations are consistent with these hypotheses.

Further evidence to support the hypothesis of rapid aggregation in this part of the cloud is seen in the vertical profiles of Snow Flux and Number Flux (Fig. 7). These quantities have been calculated by determining the number and total mass of ice particles at each height, and for each velocity bin from the Doppler spectra retrieval. The mass (or number) in each velocity bin is then multiplied by the Doppler velocity measured by the radars in order to determine the flux. Only flux values of particles >0.75 mm in diameter are shown, because the number of smaller particles cannot be reliably estimated with this combination of radars. Confidence is given to the reliability of our retrievals by the coherent structures seen in time and height (Fig. 7a,b). The vertical profile of Snow Flux and Number Flux (Fig. 7c) also supports our rapid aggregation hypothesis, because the decrease in Number Flux from 4.5 km downwards is substantially larger than the decrease in Snow Flux over the same heights. The decrease of number (flux) relative to mass (flux) is exactly what is expected from aggregation. The overall decrease of Snow Flux with height could be explained by sublimation of the ice particles in subsaturated air (included in our calculations in (5)), or through some process where large particles become significantly smaller (e.g. collisional breakup; not included in (5)). Nevertheless, these properties also support rapid aggregation in this part of the cloud.

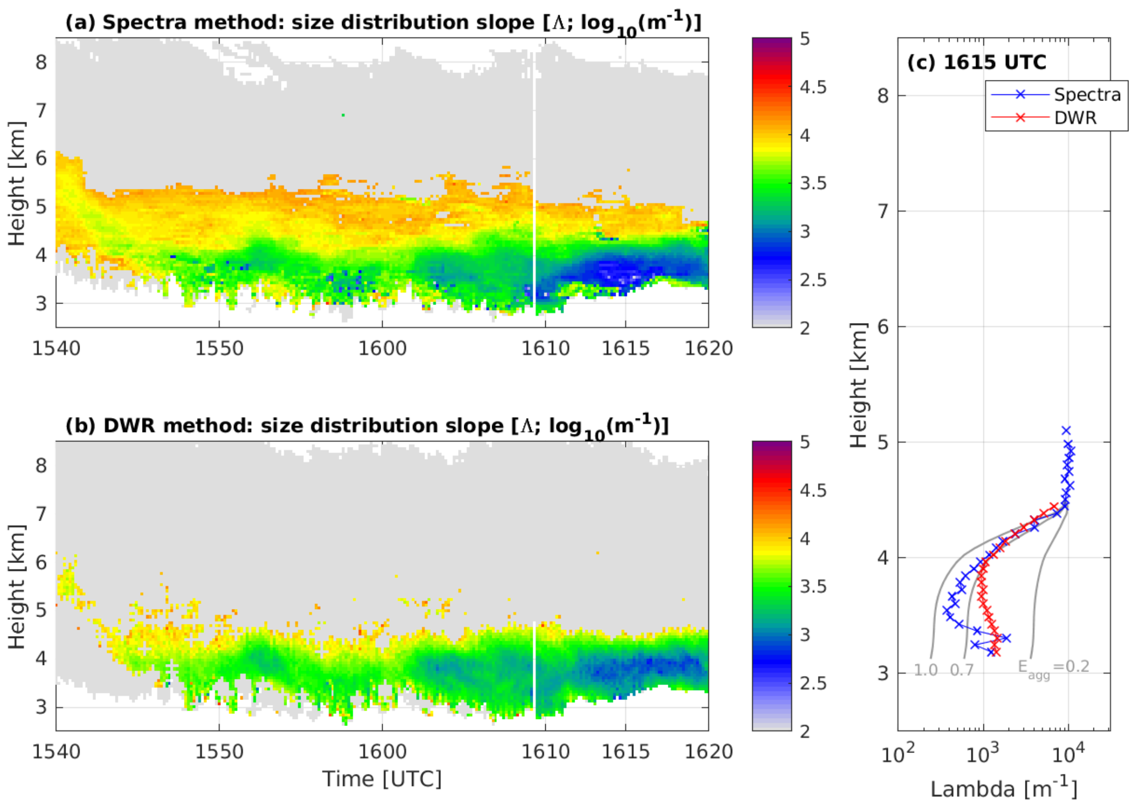

Figure 6. Time-height plots of Λ, the slope of the ice particle size distribution derived from the a) multi-wavelength Doppler spectra method and b) the dual-wavelength ratio (DWR) method. The grey regions mark areas of the cloud where no retrieval of Λ was possible. See text for details. Panel c) shows a profile of values averaged over 2 minutes centred on 1615 UTC. The grey lines show the expected changes in Λ for three different values of aggregation efficiency (1.0, 0.7, 0.2), assuming the ice particle size distribution at 4.5 km evolves due to aggregation alone. The $E_{agg} = 1.0$ CSF curve assumes a constant snow-flux from 4.5 km downwards.

6.1 Sensitivity to uncertainties in the retrieval

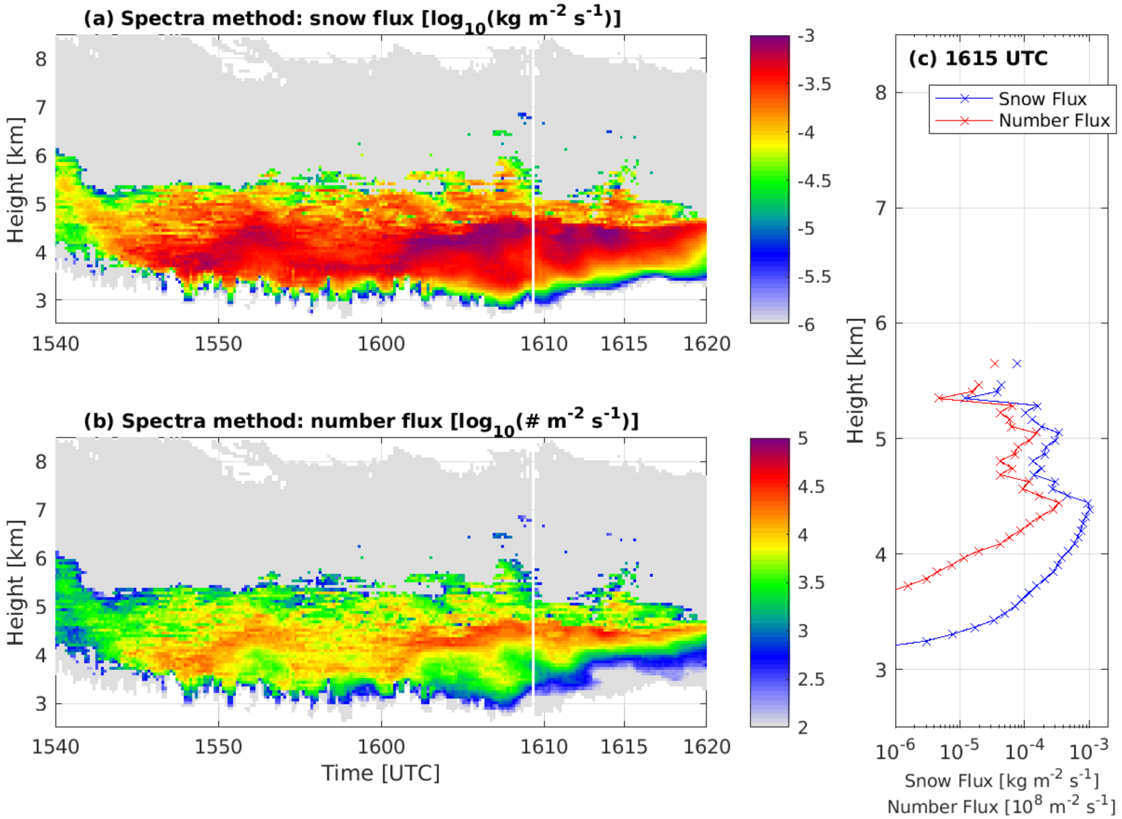

Figure 7. Time-height plots of the retrieved quantities of a) Snow flux and b) Number flux. These quantities are calculated for particles with retrieved diameter >0.75 mm only, and therefore underestimate the true snow and number flux. Panel c) shows the profile of these two quantities retrieved at 1615 UTC.

The retrieval of the properties of the ice particle size distribution is naturally sensitive to uncertainties in the input quantities. To determine to what extent our retrieval is sensitive to these uncertainties, the retrieval has been repeated with a range of different assumptions. The sensitivity analysis looks at three different aspects: 1) the impact of improperly aligning the Doppler spectra from the radars along the velocity axis; 2) the impact of improperly aligning the Doppler spectra based on reflectivity or calibration errors; and 3) the impact of using a different mass-diameter relationship in the retrieval. The details of the different sensitivity tests are given in Table 2.

Figure 8 shows how the retrieved ice particle size distribution at 4.15 km altitude and at 1615 UTC varies under the different uncertainty assumptions. There are some large variations in the maximum ice particle diameters retrieved - in particular for uncertainties related to changing the velocity (Aspect 1; blue lines) and also in the number concentration retrieved at a particular diameter can vary by an order of magnitude. However, the overall character of the size distribution is usually unchanged, and when the characteristic slope of the size distribution Λ is calculated, it is largely insensitive to the uncertainties.

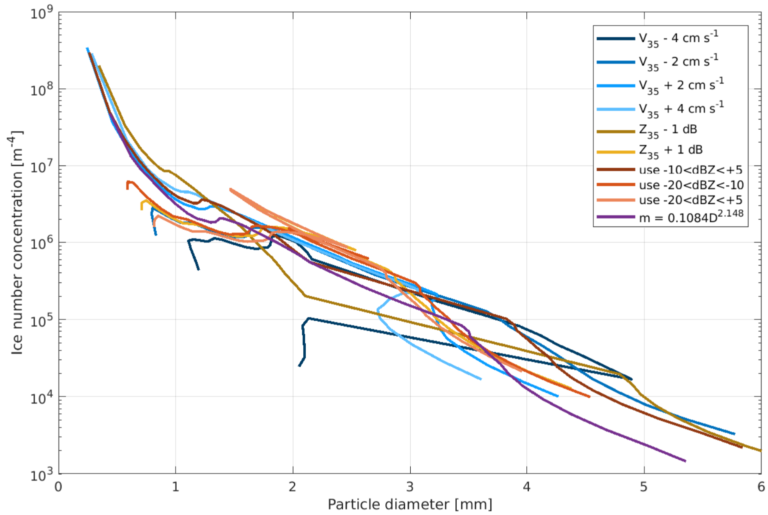

Figure 8. The ice particle size distribution at 4.15 km (equivalent to Figure 5i) under various uncertainty assumptions. See Table 2 for details about the uncertainties included in these retrievals.

This insensitivity of Λ to these uncertainties can be seen in Fig. 9. In panels a-d of this figure, the vertical profile of Λ at 1615 UTC is shown for each of the different uncertainties. This can be compared with Fig. 6c and the retrieved Λ profile from the unperturbed setup is plotted in black on panels a-d. Although there is some variation of Λ for the different uncertainty assumptions, the vertical profile continues to show rapid decreases of Λ with height down from 4.5 km, consistent with large aggregation efficiency values (Fig. 9e). The largest deviation is seen for the uncertainty where Z_{35} and sZ_{35} are reduced by 1 dB. This change results in larger Λ values at all heights due to a reduction of $\text{DWR}_{35/94}$ by 1 dB. The lower sDWR results in the retrieved particle diameters being smaller such that the largest particles have lower number concentrations and therefore a steeper slope is diagnosed. Nevertheless, the change of Λ with height for this uncertainty is also consistent with rapid aggregation. Therefore we conclude that none of the uncertainties assessed substantially change the conclusion that aggregation is likely the dominant mechanism for changing the ice particle size distribution from 4.5 km downwards between 1545 and 1620 UTC.

The estimation of the aggregation efficiency value is largely dependent on the mass-size and velocity-size relationships used, because these control the values a, b, c and d which are the main terms in (5) determining the change of Λ with height. b and d also contribute substantially to change of I_1. These values are, however, relatively well constrained. First, (5) is totally insensitive to a, because it appears only once and is cancelled out because it also contributes to the mass flux χ_f. Second, $b = 1.9$ is known for this case (Stein et al., 2015). Therefore no sensitivity exists to the choice of mass-size relationship. c and d have been estimated from the power-law fits as part of the retrieval process, and are quite well constrained within the aggregation region (Fig. 10).

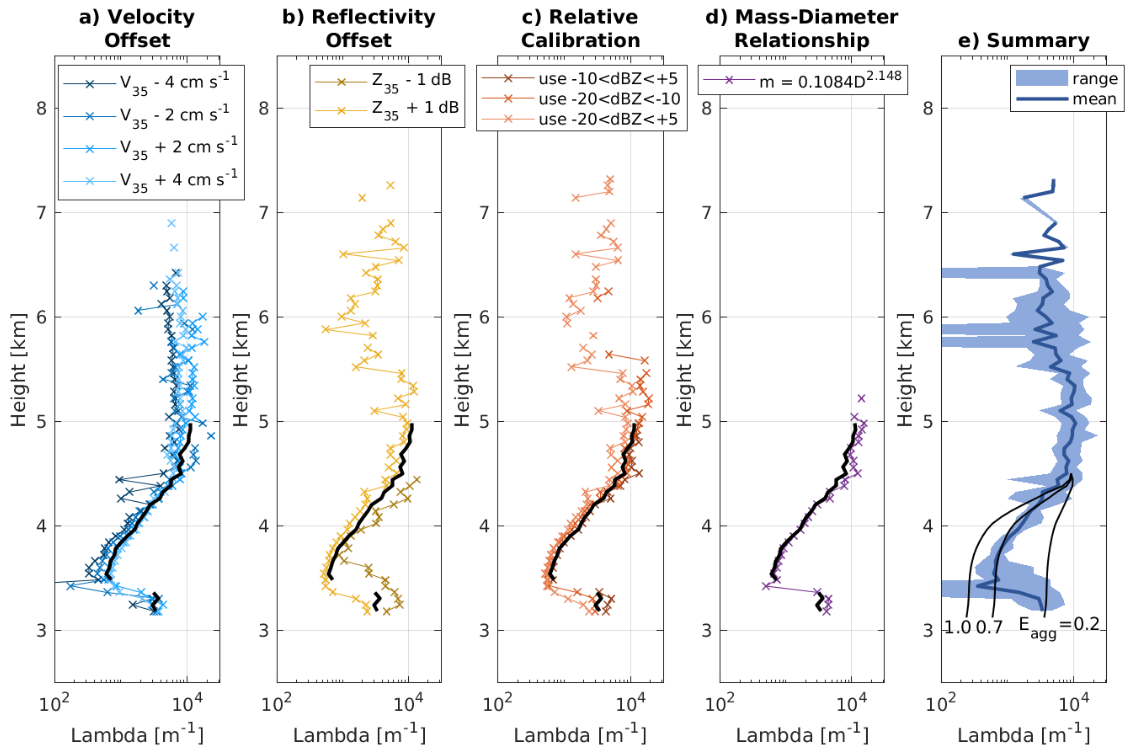

Figure 9. Panels a-d show the vertical profile of Λ at 1615 UTC (equivalent to Figure 6c) under various uncertainty assumptions. Panel e) shows the mean (solid line) and range (shaded region) as a function of height for all individual retrievals shown in panels a-d. The mean is calculated from the base-10 logarithm of the plotted values. The unperturbed retrieval is plotted on panels a-d in black for comparison. The theoretical curves for changes of Λ with height due to aggregation and starting from 4.5 km altitude for E_{agg} values of 1.0, 0.7, 0.2 are shown on panel e, as in Fig. 6c.

7 Conclusions

We have shown that the use of radar Doppler spectra data from three co-located, vertically-pointing radars at frequencies of 3, 35 and 94 GHz can produce estimates of the ice particle size distribution and be used to identify and explore processes such as aggregation. Different radar reflectivity for different radar frequencies shows evidence that there are particles present that are large enough that they are no longer within the Rayleigh scattering regime. Using the Doppler spectra from the three radars, we can determine the size and estimate the number of these ice particles.

In the case presented in this paper, we identify a region where the 35 to 94 GHz dual-wavelength ratio (DWR) increases rapidly with decreasing height, indicative that large ice particles are forming quickly. We have ruled out vapor deposition as the cause of these large particles, because that process is too slow. Similarly the rapid growth is not a result of riming because there was no evidence of significant liquid water. We therefore argue that these large particles, up to 5 mm in diameter, are

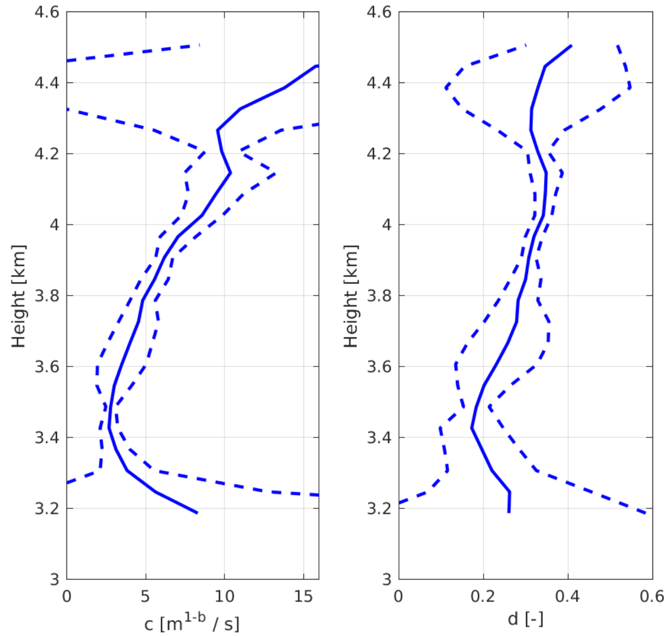

Figure 10. Vertical profiles of c and d from the power-law fits to the velocity and diameter retrievals between 1614 and 1616 UTC. The mean (solid) and the spread approximated by the standard deviation of c- and d-values in time at each height (dashed) are shown.

a result of aggregation. Our observations are consistent with theoretical calculations of ice particle size distribution evolution resulting from purely aggregation. In this case an aggregation efficiency ~~of~~ around 0.7 ~~to 1.0~~ fits the observations.

Aggregation as the cause of the rapid growth of ice particles is supported by the consistent and narrow range of heights over which this change occurs. The rapid aggregation occurs just below the region where the Doppler spectra is bi-modal, indicating the presence of small, newly-formed ice particles. It appears that the small ice particles forming at approximately 5.3 km (~~−15.4°~~ −15.4 °C), and appearing clearly in the Doppler spectra at 4.8 km (Fig. 5d), grow into dendritic ice particles at temperatures around ~~−15°~~ −15 °C and either aggregate with other similarly formed particles, or initiate aggregation with pre-existing ice particles falling through this layer. The aggregation initiated by these particles is then evident by the large particles present at 4.1 km, which could not have been formed by vapor deposition or riming.

These observations of rapid aggregation at temperatures around ~~−15°~~ −15 °C add support to cloud chamber studies (Connolly et al., 2012; Hobbs et al., 1974), which also suggest that aggregation at around ~~−15°~~ −15 °C is much more efficient that at other temperatures. The resulting changes to the ice particle size distribution through this aggregation process strongly affect many microphysical process rates (e.g. vapour deposition, sedimentation velocity, further aggregation) and therefore failure to capture these aggregation regions in models can lead to significantly errors in the simulated cloud fields. ~~Barrett et al. (2017) showed that the assumed particle size distribution is the largest single sensitivity in the model physics for~~

Table 2. Details of the changes to the retrieval input or parameters for the sensitivity testing

Aspect	Name	Description
1	$V_{35} - 4$ cm s^{-1}	Doppler spectra from 35 GHz radar shifted to the left by two velocity bins (0.04 m s^{-1})
1	$V_{35} - 2$ cm s^{-1}	Doppler spectra from 35 GHz radar shifted to the left by one velocity bin (0.02 m s^{-1})
1	$V_{35} + 2$ cm s^{-1}	Doppler spectra from 35 GHz radar shifted to the right by one velocity bin (0.02 m s^{-1})
1	$V_{35} + 4$ cm s^{-1}	Doppler spectra from 35 GHz radar shifted to the right by two velocity bins (0.04 m s^{-1})
2	$Z_{35} - 1$ dB	1 dB subtracted from Z_{35} and sZ_{35}
2	$Z_{35} + 1$ dB	1 dB added to Z_{35} and sZ_{35}
2	use $-10 < \text{dBZ} < +5$	Calibration of Z_{35} and Z_{94} to match Z_3 in regions where $-10 < Z_3 < +5$ dBZ
2	use $-20 < \text{dBZ} < -10$	Calibration of Z_{35} and Z_{94} to match Z_3 in regions where $-20 < Z_3 < -10$ dBZ
2	use $-20 < \text{dBZ} < +5$	Calibration of Z_{35} and Z_{94} to match Z_3 in regions where $-20 < Z_3 < +5$ dBZ
3	$m = 0.1048D^{2.148}$	Replaces the mass-diameter relationship from Brown and Francis (1995) with that from Heymsfield (2013) for aggregate snowflakes

~~mixed-phase altocumulus clouds. The importance of correctly simulating the ice particle size distribution has been shown in several other studies (Pinto, 1998; Harrington et al., 1999; Field et al., 2005; Morrison and Pinto, 2006; Solomon et al., 2009). Therefore understanding and correctly implementing the aggregation process in numerical models of cloud physics is important for the overall development of the cloud system.~~

This multi-wavelength Doppler spectra technique shows the ability to determine the size distribution of ice particles in large portions of ice clouds simultaneously. Previously, ice particle sizes have been determined using ice particle sizing instruments attached to aircraft, which suffer from two issues: small sample sizes and shattering of large ice particles on the instrument inlet, resulting in many small particles in the sample volume and leading to unreliable estimates of both large and small ice particle concentrations (Westbrook and Illingworth, 2009; Korolev et al., 2011). Therefore further studies of cloud microphysical structure and processes using this method are encouraged.

For the benefit of future studies, we give some advice here for achieving the best results. To achieve reliable, quantitative results from this method, the radars need to be very precisely pointed vertically. We find that mis-pointing by 0.2° is sufficient to resolve a non-negligible contribution from the horizontal wind in the Doppler spectra, which adds extra challenges to comparing the spectra from the three radars. Ideally the three radars should also have the same beamwidth; spectral broadening increases for wider beams and again makes comparing spectra from different radars more challenging, especially in the tails of the spectra where the largest DWR values are expected. Despite these challenges, we have shown that this technique enables the generation of ice particle size distributions from remote sensing data~~, which will~~. We were unable to make reliable retrievals in regions of strong turbulence (e.g. due to instability created by sublimation) because the assumption that the spectra was unchanged over the 10-second averaging window was violated. Although no low-level clouds were present on this day, the technique to cross-calibrate the radars near cloud top enables the retrieval to be performed even when (supercooled-)liquid clouds or rain are partially attenuating the radar signal at lower levels. Retrievals of this type have the potential to benefit the cloud microphysics community through both statistical sampling of clouds and aiding studies of individual processes, such as the aggregation process detailed in this paper. Further studies comparing the retrieved size distributions against data obtained from aircraft are currently being performed. ~~Our~~ One weakness of our current experimental setup ~~means~~ is that we can only size particles larger than 0.75 mm. Particles smaller than ~~this~~ 0.75 mm are in the Rayleigh scattering regime at all three wavelength and therefore their size cannot be determined. The addition of an extra ~~shorted~~ shorter wavelength (e.g. at frequencies of 150 or 220 GHz, as advocated by Battaglia et al. (2014)), would enable sizing of particles down to approximately 0.45 or 0.3 mm (for 150 and 220 GHz respectively). Such observations would provide a unique opportunity for increasing our understanding of cloud microphysics, both statistically and through process studies as demonstrated in this paper.

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