The authors would like to thank both anonymous referees for their time, their helpful comments and suggestions and their attention to all the details. We appreciate their contribution. Please find below a detailed point-by-point replies and amendments followed by the marked up manuscript. Referees comments are in blue. Citations are listed at the end of the document.

Anonymous Referee #1

Received and published: 22 August 2016

1.There is constant reference to sub- and super-saturated conditions (presumably w.r.t. ice) in the chamber, yet no RHice measurements are shown, and there is no mention if instrumentation was available to make the measurement. The lack of RH measurements creates an uncertainty in assertions that the air immediately surrounding the drops is subsaturated, saturated or supersaturated w.r.t. ice (or water for that matter).

Figures 1a, A1 have been amended to include the supersaturation data.

The text has been amended to describe the RH measurements and instrumentation as follows:

"The total humidity inside the chamber was measured by a fast chilled-mirror frost-point hygrometer (MBW, model 373LX). The supersaturations with respect to water and ice (Fig. 1a) were calculated from dew and frost points using water and ice saturation vapour pressure at the measured temperature (Buck, 1996) with deduction of the contribution from the condensed phase due to a higher temperature in the MBW sampling line".

Figure 1a amended:

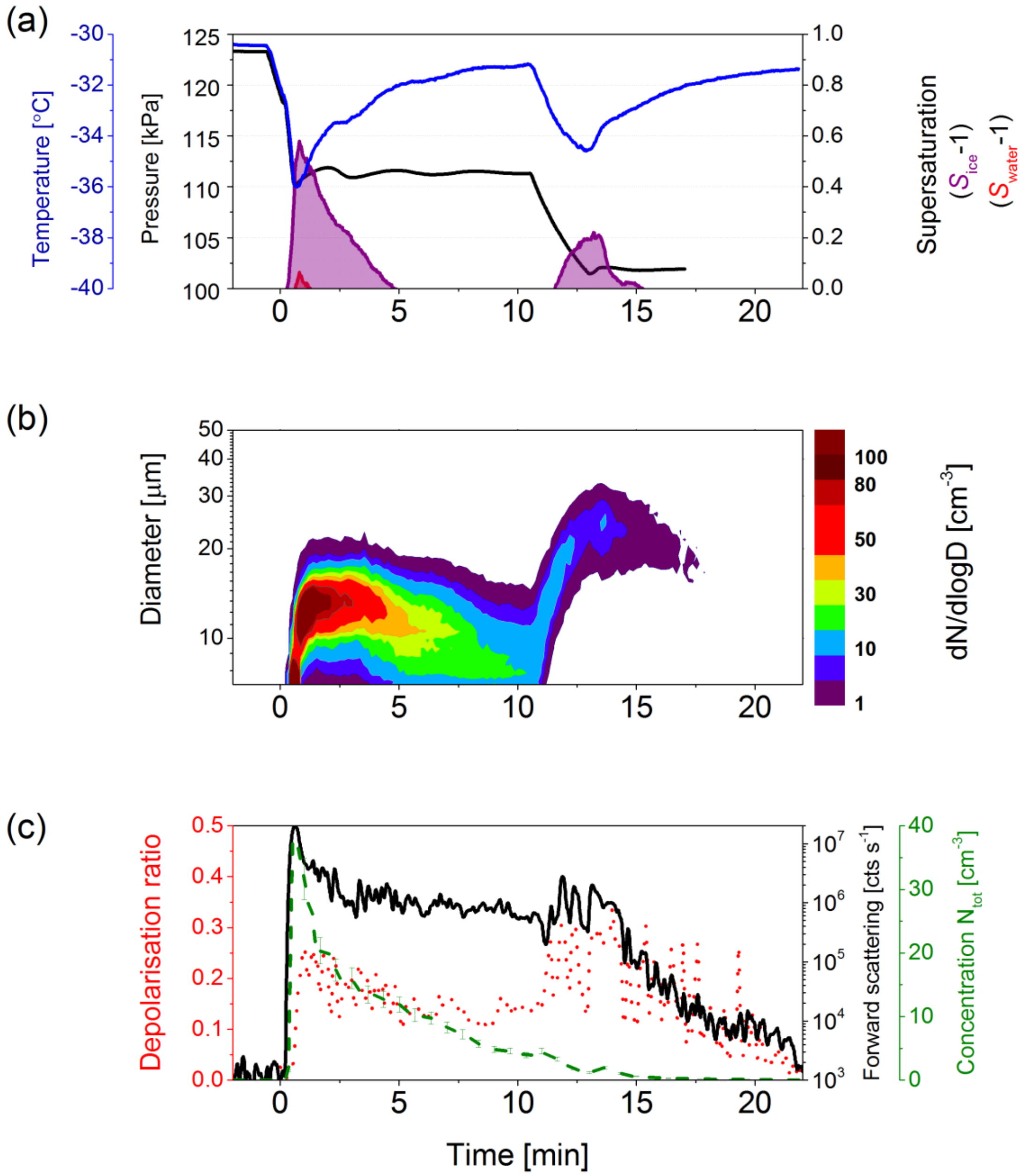
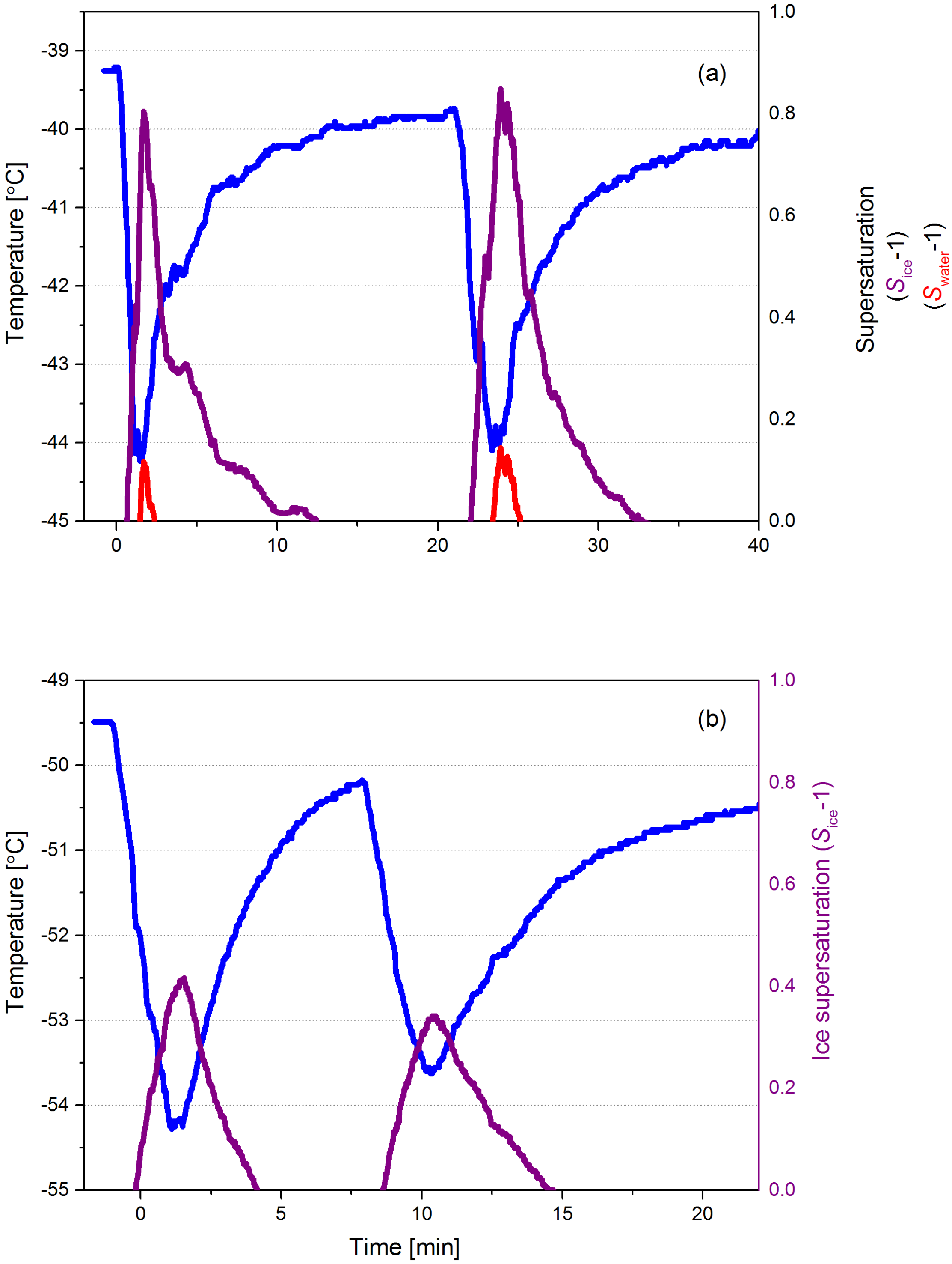


Figure A1 amended:



2. T and P are measured, but inferring that all of the water drops become ice is not supported by any measurements, but only inferred by the instruments that are being evaluated for their ability to discriminate ice and water drops. The measurements (Fig. 1) suggest rapid nucleation by CCN and formation and growth of particles following a rapid expansion in the chamber. It is assumed that these are water drops and that the drops immediately freeze. However, the measured temperature only drops to about -35.5 C, not as low as the homogeneous freezing temperature. Also, the depolarization ratio reaches a modest maximum of 0.25. There is no way to confirm if all of the water drops froze, or not. I would like to see a similar time series for an expansion conducted at the colder temperature (below – 40 C) where homogeneous freezing is assured.

- The measured temperature drops below 36 at the first step and supersaturated conditions for water are achieved (see previous point). This statement is supported by a number of previous discussions in the literature – see below;

  De Mott et al. (2011) describes the issues associated with determining the heterogeneous freezing regime: "…This challenge is a major motivation for renewed attempts to measure ice nucleation processes in general, and to design and deploy new portable systems for measuring ice nuclei (IN), the particles that are considered the only means for initiation of the ice phase at temperatures warmer than about -36°C in the atmosphere…".

  Ackerman et al. (2016): "…a greater supersaturations near cloud base (not shown), which drives greater number concentrations of ice upon homogeneous freezing of the water drops between about −35 and −36 ^{0}C…".
- Due to pressure sensitivity it was impossible to measure the activated fraction in these experiments. However, during liquid cloud expansions from another campaign (in the same chamber with the same settings), at aerosol numbers below 1000 cm^{-3}, all aerosol were activated (Hoyle et al., 2016). In our relatively high updraft speeds (see comment to Referee2 on page 25) we assume full glaciation.
- The saturation ratio with respect to water becomes sub-saturated soon after the glaciation. Therefore, it is impossible that the droplets exist throughout the experiment since they must be removed through Bergeon-Findeneisen process.
- It is not clear what is the meaning of "modest" depolarization here by the referee; however, for spherical particles the depolarization was calibrated to be zero. Before the experiments, as seen in the first instance, the concentration is at its highest value

but the depolarization signal is zero (the time gap between depolarization maximum and concentration maximum Fig 1c). The maximum depolarization is specific to the morphology and concentration of particles produced in the CLOUD chamber and is associated with any phase change and transition that presents.

Since the -50 ^{0}C experiments are complementary, as noted by Referee2, and the additional plot was required by Referee1 only as evidence to verify the phase transition, we assume that having both will satisfy each and remove the doubts.

3.There is no standard for determining whether spherical particles are water drops or ice. The PPD-2K is assumed to be capable of distinguishing spherical water from ice based on individual particle diffraction fringes, but as shown in Fig. 7, the comparison between the scattering pattern in A (water drop) and D (sublimated ice), it is not possible to unambiguously determine spherical ice from a water drop.

This discrimination is indeed an ongoing problem which we discuss in this paper and present the intercomparison between available instruments and techniques. There is no standard as yet, however, the length of the campaign and the reproducibility of the experiments in this well-controlled chamber allow us to set a variance threshold and reduce the error of discrimination. Only several examples of patterns are presented. We intentionally highlight ambiguous patterns as shown for selected periods A and D to visualize the similarity and to show that it is not always possible to derive the accurate phase of the particles. The derivation of phase is inspected first by computerised image analysis of all patterns and then visual inspection (Vochezer et al., 2016). Also, the complexity is high only on ice where submicron features may exist.

The aim of this work was to show that even in a controlled laboratory environment when the probes sample super-cooled droplets, frozen (quasi-spherical) droplets or quasi-spherical sublimating ice particles, they cannot always be discriminated with common optical instrument. This can provide a clue how well the standards of different instruments for aspherical fractions would respond in the field to derive the ice fraction. None of these instruments in this sense differentiates between ice and water but between spherical and non-spherical.

PPD diffraction patterns are more sensitive to deviation from the spherical shape and at least with manual inspection we are able to detect small deviations from the spherical shape (e.g. in pattern D one can still see “fringes”).

4. One of the conclusions stated in the Abstract is that bulk averaged path depolarisation measurements of these clouds showed higher correlation to single particle measurements at high concentration and small diameters of cloud particles. Yet, measurements of small (in this case < 7 microns) are only made by one instrument (CASPOL), and there is no way to determine why there is a (very poor) correlation (as shown in Fig. 8) and how to determine the physical significance.

Measurements of CASPOL and SIMONE are compared. The referee indicates that the PPD was not measuring below 7 microns, and only CASPOL measurements could be reported and that is indeed what we present. This is the first time that such a comparison of these instruments for different concentrations, sizes and morphologies has been demonstrated. CASPOL (an airborne instrument) and SIMONE measurements are compared with the PPD in figures (1c, 3, 6) and this comparison is now presented for completeness.

These results are important for complementary bulk and single particle measurements to understand how instrument responses are affected by size and concentration in different environments. Both instruments measure a polarisation product therefore this comparison is essential. Thresholds derived from this comparison will be added as follows:

“Based on our analysis, ensemble depolarisation measurements of cloud particles at concentrations above 20 cm^{-3}, sizes below 15 μm and certain atmospheric conditions can be comparable to single particle polarisation airborne measurements.”

The statement in Section 3.4 lines 33 – 34 that the correlation in Fig. 8 is surprisingly reasonable leaves this reviewer bewildered. It looks to me like the correlation is terrible. The (max) R2 value of 0.35 in regions with small particles at high concentrations (where there is no way of actually knowing the shape of the particles) is nothing to brag about, and R2 = 0.01 in regions with low particle concentration is pitiful.

The referee must be referring to lines 23—24. This sentence is changed now to: “Generally, we have found that higher correlation is observed (Fig. 8a, 8b) between the different instruments in cases with high concentration and small diameters of cloud particles (R^2 ~ 0.35) and almost no correlation (R^2 ~ 0.01) in cases with low concentrations and larger sizes of cloud particles.

"where there is no way of actually knowing the shape of the particles" – The optical shape of the particles at "high concentration" mentioned in this paper can be easily derived from polarisation i.e. spherical/aspherical, the main problem here is phase derivation from the shape which could be ambiguous.

5. The 3V-CPI is the only instrument that provides actual images of these particles. Even though the CPI pixel size resolution is not optimum for resolving the shape of these small particles, the manuscript needs to show more images of particles. Specifically, show images of the water drops prior to freezing. Also, there is mention of columnar shapes identified by the PPD-2K, but no CPI images. Please show the CPI images that correspond with the PPD-2K derived columns.

Water drops prior to freezing rarely reach the lower threshold of detection (e,g Fig. 1) and even if they do the conditions for imaging such as DOF, dead time etc. (Connolly et al, 2007) as well as sampling location, limit the number of images to very few, which may make them statistically unrepresentative depending on the environment. Additionally, it is not clear what would be the scientific value of the comparison between low resolution spherical water images to almost the same spherical images of frozen droplets.

No CPI images are available for the columnar shapes since in this case they were below the size threshold for shape discrimination (e.g. Fig. 3). In other CLOUD experiments with larger sizes generated which are beyond the scope of this paper, columnar shapes were observed with the CPI (e.g. Nichman et al., 2016).

"The pixel size resolution is not optimum" in my opinion this is an understatement , we are measuring on the edge of the detection and resolution ranges of all currently available instruments and demonstrate the limitations of their discrimination abilities in these conditions.

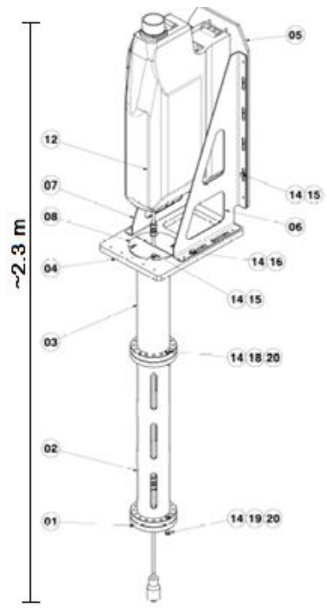
5. I don't understand the CPI measurements in Fig. 6. How are the gray squares calculated? Why are there multiple overlapping measurements at the same point in time? If each point

represents an individual image analysis, then why weren't the other single particle measurements processed in this manner. Why are there not more CPI measurements in Figs. 6b,c?

As mentioned in the text and in the caption the grey squares are fractions of particles classified as non-round out of total population in every second. The time scale of minutes and the size of the markers may cause a confusion of several points at the same time. We have smoothed the CPI data, reduced the size of the marker and changed the colour to magenta for clarity. In Fig. 6 b,c the majority of particles haven't reached the detection range of the 3V-CPI (see Fig. 3).

6. There is no description of how the instruments were operated. Were the instruments installed in the cloud chamber? Was cloud air exhausted through the sample volume of the probes? Were the probes aspirated? Etc. There also needs to be more description of how the instruments were operated and how the measurements were processed.
It is not straightforward how to measure the sample volume of instruments used to measure particle size distributions from a cloud chamber. How were the size distributions computed? The agreement in the size distributions shown in Fig. 2 is poor, often differing by an order of magnitude. How does this affect the results reported in the paper?

The chamber air was sampled through the sample volume of the probe at a laminar flow with air speed of about 4 m s^{-1} for CASPOL and 3V-CPI. CASPOL was measuring through a horizontal sampling line. PPD had a sample flow of 5 lpm and a sheath flow of 2 lpm (Additional information regarding chamber air volume probing can be found in Jarvinen et al., 2016c; Duplissy et al., 2016). A diagram showing the probe positions around the chamber was added as a Supplementary material. Additional details were included in the description.
A technical drawing showing the 3V-CPI instrument and how it was attached to the chamber using a vertical sampling line is shown below. This can be included in the supplementary material if the Editor recommends, but we feel this level is not required and was not required in the cited references using a similar instrumental setup.



The size distributions presented are an average over 30 s during a stable period of the expansion selected to minimize chamber mixing artefacts. Undercounting by the PPD even at low concentrations could result from its sampling location. The discrepancies in concentration indicate measurement bias but do not affect the polarization properties and asphericity intercomparison which we report, since there is no coincidence problem at these concentrations as showed on page 18 in this response.

Overall, I am not sure what the takeaway messages are from this paper. None of the instruments tested are capable of unambiguously distinguishing ice from water.

This is exactly the message. None of the instruments, including many not examined in this paper are able to unambiguously detect the phase of small quasi-spherical cloud particles. While reports of the ice phase are more cautious and often underestimate ice fraction by including only aspherical shapes, reports of droplet concentration normally do not take into account spherical ice (e.g. sublimated) thus overestimating the liquid fraction. Here we highlight this issue and test one possible method for phase discrimination of small quasi sphericals.

The main point in this paper is the comparison of aspherical fractions from different instruments; analysis of the complexity is presented as one potential pathway to explain the ambiguity. However, the main goal is to show that the phase of the small quasi-sphericals cannot be unambiguously classified with some of the common techniques.

"Complexity" is discussed but never really defined, except to hypothesize that it is a "frost" layer.

The surface anomalies which are represented in the measured complexity are mentioned on page 9, line 38: "Although these anomalies, like roughness and stepped hollowness of the crystal, do not significantly contribute to the mass distribution, they can significantly alter the light scattering properties of the ice crystals…".

Frost layer was already mentioned by Jarvinen et al. (2016a), cited, and earlier. It is used here to concisely describe the features mentioned above and to proceed with the terminology in these complementary experiments.

Based on the PPD-2K diffraction images, the instrument can show the difference between a spherical particle and a particle that is irregular in shape or has some surface "complexity", but there is no convincing explanation of how to apply this information quantitatively.

The complexity parameter was introduced as supporting information; it was never intended to promote quantitative application of this complexity.

Page 12, line 29-30: This complexity measurement can be potentially calibrated in future experiments to derive the complex fraction of ice particles.

Proper calibration would provide the threshold for discrimination of the complex ice crystals, similar to the complexity threshold used in Schnaiter et al. (2016) for SID-3 and Jarvinen et al. (2016a).

The k value is mentioned, and in other papers there are examples of diffraction patterns from analogs and other shapes, but there is no comparison with high-resolution images of actual ice particles. The images from the PHIPS-HALO instrument in Schnaiter et al. (2016) do not have adequate resolution to provide useful information, except to distinguish columns from quasi-round particles. After looking at the diffraction patterns in Schnaiter et al. (2016) I cannot tell the difference between a distorted (analog) scattering pattern and one in this manuscript that is labeled as having surface complexity. There also appears to be no additional information on how well the diffraction patterns correlate with actual high-resolution images of ice particlesin Vochezer et al. (2016). Ideally, an instrument capable of imaging particles with much higher optical resolution than the CPI should be used to compare with the PPD-2K. Could ice particles be captured on a cooled slide, placed in a cold box and photographed under a microscope?

The reviewer is right that high-resolution methods are needed to image surface features or small-scale complexities, such methods however are only available in microscope scans that require placing the particles into the microscope, i.e. they are no-longer airborne. During the continuous CLOUD campaign, collection on slides at low air speed and examination in a suitable remote facility (not available at CERN) has its own drawbacks and was not feasible. For such particles one cannot measure the light scattering properties and it is not completely clear how the sample is affected by the beam in a (usually evacuated) microscope scan chamber. This recommendation will be added to conclusions.

It is not clear which figure the referee refers to as "distorted (analog) scattering pattern" whether he tries to compare PPD-2K scattering pattern in our experiment to Figure A1 of ice analogue aggregate with rough surface residing on a polycarbonate window or Figure B1 in section **Modeling SID-3** scattering patterns in Schnaiter et al. 2016. Either way, these scattering patterns cannot be directly compared between different papers, with different calibrations and instruments:

First, we did not have any aggregation (which can be identified by the CCD imaging probe) in our short time-scale cloud experiments which simplifies the classification of patterns.

Secondly, the distorted spherical ice model particles with different deformation parameters (Fig. B1 in Shnaiter et al. 2016) show how the surface deformation setting in the model can affect the roughness parameter for SID-3.

Nonetheless, a comparative analysis of the patterns can be achieved within the same experiment as reported in our paper.

Even though the CPI only has adequate resolution to distinguish round, quasi-round and columnar shapes for particles $\geq$ 30 microns, I would still like to see a comparison between CPI images and PPD-2K diffraction patterns of the various particle shapes that are mentioned in the manuscript.

Already answered in point 5: No CPI images are available for the columnar shapes since in this case they were below the size threshold for detection (e.g. Fig. 3). In other CLOUD experiments with larger sizes which are beyond the scope of this paper, columnar shapes were observed with the CPI (e.g. Nichman et al., 2016).

The SID family of instruments (including the PPD-2K) provide interesting and potentially useful measurements, but the quantitative utilization of these measurements in mixed-phase and in cirrus clouds with a combination of growing and sublimating particles is not clear. Measurements have shown that a substantial fraction of false irregulars are seen in all-water clouds (i.e., Johnson et al. 2014 JAS). Yes, certain pristine shapes can be identified: perfect spheres, column shapes and possibly hex shapes, but the large majority of ice particles in cirrus are irregular. How are these particles quantified?

The SID family is well known to have serious coincidence problems especially at high concentrations, the more so in mixed-phase cloud for phase discrimination, especially in real atmospheric environment. In our study every deviation from the spherical shape is quantified as aspherical, therefore plates, columns and other irregular habits would be "aspherical", their fraction from the total is represented in percent. The title of the paper "Intercomparison study and optical asphericity measurements of small ice particles…" states the goal of asphericity measurements. Habit classification is beyond the scope of this paper.
The referee must be referring to Johnson et al 2014 JTECH: "…Remaining uncertainties in the sensing volume and the volume over which coincidence of particles occurred, result in the data being used here in a qualitative manner to identify the presence of ice, and its habits and sizes". The SID2-H that has a slightly different measurement principle than PPD-2K has poorer spatial resolution. It is not clear if coincidence occurs how the shape of a single particle can be detected with subsequent accurate phase derivation. This could explain the high number of irregulars. This detection of false irregulars occurs in all-water and mixed-phase clouds of high number concentration. With the newer generation of SID-family instruments the coincidence images can be identified, although currently only through manual inspection. Although larger particles are often irregular; in the small size range, spherical particles are frequently reported.

The ability of the CASPOL to quantitatively distinguish water and ice is not demonstrated at all. The results vary with both particle size and concentration, leaving one to wonder what it is really measuring. There is good qualitative agreement with the PPD-

2K in estimating asphericity in Fig. 6a, but no agreement in Figs. 6b and 6c. What is the explanation for this?

The agreement in figure 6a is quantitative, based on measured polarisation ratios and clustering analysis as previously shown by Nichman et al., 2016. One possible explanation for low agreement is that PPD is more sensitive to subtle features as we showed. Another possible explanation is described in section 3.1.2.

In the cirrus temperatures the ice particles were smaller and we had a mode that reached the PPD size range whereas in the droplet freezing experiments these smaller ice particles were in lower concentration. Also, the sphericity of the particles in these two regimes is different.

Similar discrepancies are seen in Jarvinen et al., (2016a), it seems that in those cases the majority of small particles were not detected by the PPD-2K but were detected by both CASDPOLs (a slightly different versions than our CASPOL). For the homogeneous case (Fig.4 in Jarvinen et al., 2016a) only about 37% of the particles detected by CASDPOL were in the PPD size range (personal communication) all the other were smaller hence occupying the places in the 292 PBP bins and then filtered out, increasing the margin of error for this comparison and inducing discrepancy. This is why it was important for us to show the PSDs of the two instruments which help to interpret the measurements and explain the discrepancies.

Specific Comments:

P. 2 Lines 19 – 20: I disagree. Shape is used more often than scattering intensity in mixed-phase clouds, and arguably more reliably. In many cases in mixed-phase (i.e., water saturated) clouds, ice particles rapidly grow to sizes where they can be distinguished from water drops using CPI imagery (see Lawson et al. 2015 – JAS).

"For the detection of particle shape and structure, the scattering intensity of single particles is most commonly used."

- Referee 1 argues that "shape" is used more often than scattering intensity. It is not clear what "shape" technique he refers to. Is it Imaging/ shadowgraphs/ diffraction/ holography/ scattering? all can detect shape in one way or another.
- If it is imaging that is used more often than scattering, Lawson et al (2015)-JAS demonstrate the opposite, most instruments are scattering probes (FCDP, CDP, FFSSP) vs. Imaging (CPI) and Shadowgraph (2DS, HVPS-3).

- Our paper focuses on small particles < 50 micron. Imaging becomes less useful close to the lower boundary of the geometric scattering, the resolution is inadequate.
- Text was amended to "…commonly used..".

P. 2 Lines 32 – 32: The measurement of particles smaller than 50 micron using the FSSP were contaminated with shattering. Delete this reference.

"Lawson et al., (2006) reported that particles < 50 micron account for 99 % of the total number concentration, 69 % of the shortwave extinction, and 40 % of the mass in mid-latitude cirrus".

- I'll add that there was shattering, nonetheless the referee suggests the measurements were contaminated with shattering, indicating that shattering alone could not be responsible for 99% of the total number concentration. This citation is important to demonstrate that small particles are ubiquitous:
  "Moreover, Lawson et al., (2006a) reported that particles < 50 μm account for 99 % of the total number concentration, 69 % of the shortwave extinction, and 40 % of the mass in mid-latitude cirrus, however, these measurements were contaminated with particle shattering".
- Luebke et al., (2016) defined all <10 micron particles as spheres (derived from an inspection of the sphericity of the ice crystals, which shows that there are many spherical ice particles present during the campaign, especially at the smaller sizes).

P. 4 Lines 1 – 3: "We then use the asphericity to determine the ice fraction in a cloud by prescribing an aspherical shape for all the ice particles, and hence assume that ice fraction is equivalent to an aspherical fraction." As discussed above, using the measurements presented in this manuscript, there is no way to unambiguously determine if asphericity explicitly distinguishes ice particles from water drops. This statement needs to be modified or deleted and then explained later in the text after it is understood that using asphericity is an estimate of ice fraction that is not well quantified under all conditions.

Text amended: "Asphericity is often used to determine the ice fraction in a cloud by prescribing an aspherical shape for all the ice particles, and hence assuming that ice fraction is equivalent to an aspherical fraction, this practice is tested in our experiments".

P. 4 Lines 10 – 14: This statement appears to be contradictory. If LWC is independent of updraft velocity, but stronger updrafts produce a higher concentration of smaller drops, which then freeze, how is IWC increased in stronger updrafts? This appears to violate conservation of mass.

This is clearly an incorrect citation, thank you and Referee2 for bringing it to our attention. Text amended:

"This is clearly shown by Ackerman et al. (2015) where ice particle mass distributions in homogeneous freezing for stronger updrafts produce substantially smaller ice particles".

P. 5 Line 6: 100 per cc is not necessarily a low concentration. Simulations now show that coincidence occurs at this concentration with the CASPOL and multiple scattering will occur in ensemble measurements. Please qualify this statement (and not by using 1980's references to the FSSP).

"At the beginning of most experiments, we generated low concentrations (~ 100 cm^{-3}) of sulphuric acid aerosol".

- This concentration was measured with aerosol instruments such as SMPS and CPCs (Hoyle et al., 2016). The CPC will be mentioned in the text:
  "At the beginning of most experiments, we generated low concentrations (~ 100 $cm^{-3)}$ of sulphuric acid aerosol counted with a condensation particle counter (CPC, TSI model 3010)".
- 100 per cc of CCN is not necessarily the concentration of ice particles which are measured in these experiments. Lance et al., (2012) concluded a significant oversizing and undercounting at ambient droplet concentrations of 500 cm^{-3}, however no significant coincidence is seen from the plots at concentration below 100 cm^{-3}. A citation of reported coincidence simulations for CASPOL at such low concentration could be useful.

P. 5 Lines 26 – 30: Please show some quantitative evidence that 10-5 asphericity threshold actually applies to ice/water discrimination. Otherwise, please state that this is a subjective value based on visual analysis of the scattering pattern. Referencing Vochezer et al. (2016) is not sufficient.

The Variance threshold is applied for aspherical discrimination and not necessarily for phase (ice/water) discrimination as we show in this paper (e.g Fig 6a where spherical ice is misclassified).

This threshold was determined in laboratory experiments at the AIDA chamber with the same instrument Vochezer et al. (2016) and was further used in the study of Järvinen et al. (2016a) with the same instrument, where particles below this threshold were classified as spherical. The typical value for water droplets was between 2E-6 and 1E-5, depending on the droplet size, while the typical value for columnar ice particles was around 1E-1 (Jarvinen et al., 2016a) - we added the citation of this paper in this section.

Text added: “This threshold is used based on the visual inspection of diffraction patterns of ice and droplets in chamber experiments”.

P. 6 Lines 1: Detection of a bulk cloud phase is meaningless unless the cloud is allwater (T > 0 C), or known to be all-ice (i.e, colder than – 40 C). There is no quantitative information published (yet) on bulk measurements of the ice fraction in mixed-phase.

Mixed-phase clouds in particular remain one of the greatest sources of uncertainties in the modelling of the Arctic response to climate change due to an inaccurate representation of their variability and their quantification. The lifetime of mixed phase clouds (MPS), particularly in Arctic latitudes, is related to the number concentration of ice particles in them. If concentrations are low then cloud dynamics is able to sustain mixed phase conditions, however once concentrations increase beyond a threshold then MPS tend to glaciate rapidly via the WBF mechanism. For this reason high ice concentrations are normally not observed in most Arctic MPS where droplet concentrations are also small, (McFarquhar et al. 2011; Jackson et al. 2012). Knowledge of this transition threshold is critical therefore for models, however ice formation in Arctic MPS appears to be generally over-predicted by models at present due to over representation of ice formation processes. This leads to modelled cloud fractions that are often significantly less than those observed (de Boer et al. 2014) with widespread MPS decks common. However, Arctic MPS in summertime are more likely to be multi-layered and develop precipitation (Barrie 1986; Curry et al. 1988) partly due to the presence of secondary ice production processes. This is an area of ongoing research but highlights the need to discriminate ice and water contents for model diagnosis.

Although bulk measurements of mixed phase clouds are meaningless, as the referee claims, we believe it is highly important for observation of phase transition. Constant efforts are made to measure and report mixed phase clouds using space, ground and airborne remote sensing (averaged bulk) in various campaigns (e.g. Zhang et al., 2014) for the reasons discussed above. Chamber measurements and inter comparisons of this kind could shed more light on effects of particle alignment, shape, concentration, and sizes on these measurements as well as quantifying the accuracy with which in situ measurements can be determined.

P. 6 3V-CPI: The references in this section are terrible, misleading and in one case unavailable. The 2D-S portion of the 3V-CPI should be referenced by Lawson et al. (2006) – JTech. The CPI portion of the 3V-CPI should be referenced by Lawson et al. (2001). Lawson et al. (2003) should be deleted. The Heymsfield et al. (2010) reference does not show particle habit classification schemes. This reference should be replaced by, for example, Lawson et al. (2006) – JAMC; Um and McFarquhar (2009) – QJRMS; Lindqvist et al. (2012) – JGR

Of course the CPI technology is being constantly updated and analysis techniques improved. The CPI referenced in Lawson et al. (2001) while generically similar to that used here is technically inferior to the current version (sometimes referred to as CPI 2.0 with much higher capture rate), but we are happy to use the additional references suggested. Text amended.

Section 3.1.1: As explained above, there are way too many assumptions about what is happening during the first rapid expansion and for a few seconds or minutes afterward. How do we know that all of the drops froze instantaneously? Could there be a mixed phase cloud and Bergeron process occurring after the rapid expansion?

Ice and water supersaturation plots were added to support our statements. The mixed phase cloud may exist only during the expansion, however after the fast expansion all droplets are frozen, in water subsaturated conditions. The “assumptions” are also supported by the several cited references of previous cloud chamber investigations, which we refer the reader to.

P. 8 Line 16: How do you know there was no coincidence?

Using formula 1c from Vochezer et al., (2016) for coincidence probability:

$$P(x > 1, \lambda) = 1 - (1 + \lambda) \cdot e^{-\lambda}$$

We get that most of the time the probability for coincidence was negligible, below 1 %:

220 cm^{-3} → 0.21 %

160 cm^{-3} → 0.11 %

110 cm^{-3} → 0.05 %

150 cm^{-3} → 0.098 %

750 cm^{-3} → 2.18 %

A coincident sampling probability of 1 % is reached at particle number concentrations of 495 cm^{-3} for the PPD-2K.

P. 12 Lines 1 – 9: There are several assumptions and generalizations in these lines that need to be deleted based on previous arguments in this review.

Changed to:

Aspherical fraction derived from CASPOL data can be compared to other instruments with higher confidence when the PSD is fully covered by the overlapped size and concentration range of the instruments with sufficient number of particles for aspherical fraction derivation and low standard deviation. Such a comparison can reveal the true ice fraction, however, high concentration may cause coincidence and misestimation of the ice fraction.

The comparison of remote sensing and PBP measurements is not a straightforward process (i.e. bulk vs. single particle and single complexity vs mixed-complexity ensembles of particles). Many single particle and ensemble measurements laboratory techniques in particular have proven difficult to adopt when translated to real atmospheric environments. These techniques often provide complementary data rather than comparable data (Lynch, 2001) and research in this area continues. Based on our analysis, ensemble depolarisation measurements of cloud particles at concentrations above 20 cm^{-3}, sizes below 15 µm and certain atmospheric conditions can be comparable to single particle polarisation airborne measurements.

Recent efforts to classify clouds (e.g. Krämer et al, 2016; Luebke et al, 2016) by their microphysical properties did not account for particle morphology in their study; the morphology of small ice of in situ cirrus or of liquid origin clouds in the first steps of homogeneous freezing may affect the scattering properties and hence the solar radiation budget. According to the findings in this paper, the complexity might be present however undetectable by some instruments at lower temperatures or small sizes (<7 µm). It may increase scattering and thus intensify cooling while the lack of complexity of liquid origin clouds in warmer subzero temperature may allow more efficient warming. However, this should be further investigated.

Conclusions: This also needs to be re-written to tone down all of the claims that are not substantiated.

Changed to:

We have presented an instrumental setup for combined single cloud particle and ensemble measurements for assessment of the relative optical ice and liquid responses in each case. The results were used to determine the asphericity and small-scale complexity evolution during adiabatic expansion, sublimation and regrowth as well as for potential impact on phase discrimination. We report observations of super-cooled and frozen droplets, small ice habits and spheroids in a series of CLOUD chamber experiments at –30, –40 and –50 °C.

We have shown that the small quasi-spherical ice particles produced in the sublimation process exhibit a similar optical behaviour to that of water droplets in the PPD-2K variance analysis and in the CASPOL polarisation analysis for high PSD overlap at –30 °C. The analysis of the scattering patterns shows the similarity of the spherical states and the difficulty in applying automatic phase discrimination. Therefore, observations of small spheroids (< 40 µm) in sub-saturated conditions might be highly ambiguous. These results indicate that small quasi-spherical ice misclassification might similarly concern numerous optical instruments, impactors and other probes that were not examined here. Nonetheless, the scattering patterns differ for quasi-spherical ice and water due to small deviations from sphericity. An increase of resolution in future versions of the optical instruments might amplify this discrimination and reveal additional subtle features. . In atmospheric measurements, small particle detection is often contaminated by shattering therefore coincidence should be addressed before any comparison can be made.

We have shown a chamber simulation of small-scale complexity evolution on a frozen droplet during an updraft and in sub-saturated conditions. In regions with high concentration of small cloud particles (< 40 µm), the observed differences in morphology will affect the observed radiative properties, growth mechanisms, aggregation and charging in clouds. The aspherical fraction detected by the PPD-2K could be described with a high degree of small-scale complexity which was undetectable by the other instruments. Future efforts should aim on calibration of the scattering pattern analysis thresholds and interpretation of the reported complexity to derive the complex fraction of ice particles. It would be useful to collect and analyse the particles offline with high resolution microscopy to confirm the airborne particle measurements. It should be highlighted that the phase of small spherical particles with low complexity cannot be unambiguously defined with any of these instruments and the complexity of ice particles smaller than 7 µm remains unclear.

We have presented polarisation measurements of airborne and laboratory-instruments in an expansion chamber. We conclude that in these simulated atmospheric conditions the polarisation and depolarisation signal from frozen droplets have higher correlation at higher concentrations of small particles and can be comparable above certain concentration and size thresholds. These findings and the derived instrumental differences may be used in the interpretation of atmospheric measurements of frozen droplets from remote and in situ combined campaigns as well as a pathway for further research and development of these instruments.

BTW – the measurements presented in this manuscript extend out to particle sizes of about 30 microns. On line 26 and another place in the manuscript
the claim is that the results are valid out to 60 microns. Where does this come from?

60 micron is the number that often appears in the literature as the upper threshold for small ice category, and imaging becomes more accurate above this threshold.
The results are valid out to 40 micron, text was amended.

Anonymous Referee #2

Received and published: 31 August 2016

There is a great need to understand this type of measurement and so the aims of this paper are highly relevant at this time. What the work does do is to highlight the difficulty, even in the controlled conditions in the cloud chamber, of making the distinction between spherical ice and liquid particles, and this is valuable, especially when presented for a relatively new instrument like PPD2K.

The implications of these difficulties are not properly explored though, and if this were expanded the study would benefit. There is no real path forwards presented and little guidance as to where and when each type of measurement can offer clear advantages.

- Conclusion section was edited to include future efforts (see response to Referee1 p.19)
- Implications section amended, text added:
  Recent efforts to classify clouds (e.g. Krämer et al, 2016; Luebke et al, 2016) by their microphysical properties did not account for particle morphology in their study; the morphology of small ice of in situ cirrus or of liquid origin clouds in the first steps of homogeneous freezing may affect the scattering properties and hence the solar radiation budget. According to the findings in this paper, the complexity might be present however undetectable by some instruments at lower temperatures or small sizes (<7 µm). It may increase scattering and thus intensify cooling while the lack of complexity of liquid origin clouds in warmer subzero temperature may allow more efficient warming. However, this should be further investigated.

The PPD2K seems to offer benefits, but the chance to fully exploit this measurement is not taken here, for example - the calibration of surface complexity has not yet been performed. Work to fully characterise the small scale complexity would help, but the instruments used for comparison (CASPOL, 3V-CPI), as stated in the work, do not have sufficient resolution, which makes the aim difficult.

Nonetheless, these results complement previous homogeneous freezing intercomparison from the AIDA chamber (Jarvinen et al., 2016a) by comparing additional instruments and adding more atmospheric scenarios.

The work is somewhat over ambitious in its claims and is rather unfocussed. Is this a comparison between different ice formation situations (liquid vs. in situ), or is it a single particle instrument comparison, or is it a single particle vs. bulk averaged measurements comparison? It is in part all of these, but with the result that not one area is explored in that much depth. If this work is to be a comparison of different ice formation scenarios then the details of the in situ cases are hidden away in the supplementary material somewhat. If the work is to be a detailed instrument comparison then there needs to be more discussion about the instruments and processing, e.g. phase discrimination and techniques and thresholds.

As the title suggests we present an intercomparison study and optical asphericity measurements of small ice particles by common detection instruments in various ambient conditions.

More details of the in situ cases were added e.g. supersaturation plots, sampling.

Phase discrimination thresholds are explained in previous papers Nichman et al., 2016 and Jarvinen et al., 2016a, b.

More technical details of sampling were also added for each instrument section.

The supplementary information was moved to appendix A to be readily available in the paper.

Both the abstract and conclusions sections should be thinned out to contain only the firm statements of work done and the robust conclusions or the claims supported by stronger evidence. If this is done and the following recommendations are accounted for then the work should be promoted to AMT.

Stronger evidence is provided in the supersaturation plots.

Abstract and conclusions amended.

Specific sections

There is no mention of small hexagonal ice after the abstract.

We do mention column habits which are more common at cirrus simulations and by definition are hexagonal. This is just to demonstrate the presence of other habits which can be discriminated. The main focus is on spherical /aspherical discrimination rather than habit classification. The aspherical fraction includes plates, columns and other irregulars (also mentioned in response to Referee 1 (p. 12) about the SID family of instruments).

2.1 Section 1 - Introduction

There is no mention here, or elsewhere, as to on what scale the small-scale complexity is expected to be present for atmospherically relevant particles. This is required for discussion of the PPD2K and comparison with the resolution of probes as it is mentioned often in the text.

Regarding the dimensions of the complexity,

Page 10, line 4: "This instrument is sensitive to features that are on the order of the wavelength used, 532 nm".

Page 10, line 10: "However we should emphasize that at present it is not possible to quantitatively relate this value to an actual degree of complexity or surface uniformity of the particle".

Text was added to the introduction:

"Gayet et al., (2011) reported that in a deeply rough surface the mean free path length between two subsequent inclusions was equal to 15% of the diameter of the circle circumscribing the hexagonal facet of an ice crystal".

As to the atmospheric scale of surface complexity: Gayet et al. (2011) suggest that while simple pristine crystals are uncommon, particles with imperfect or complex shapes (shape defaults, roughness, inclusions, . . .) are prevalent.

It is not fully clear yet what is the exact extent of this small scale complexity in the atmosphere, however, it is clear both from modelling and experiments (Gayet et al., 2011; Scherbakov et al. 2006; Baran and Labonnote, 2007; Baran 2012) that it can significantly alter light scattering.

Mentioning the sizes and concentrations ranges of particles in the clouds (line 24) would be helpful here, especially when referring to remote sensing limitations (size / wavelength dependant).

Text added to section 1: "Droplet concentration in clouds normally varies between several tens to hundreds cm^{-3} while typical ice crystals concentration is normally a few particles L^{-1}

but can reach ~100 L^{-1} in old clouds (Wallace and Hobbs, 2006). The diameter of single droplet or ice crystal is normally in the super-micron range and can reach several hundred µm.”.

2.2 Section 2 - Methodology

The paper tends to present the work in terms of the -30 degree C case, the liquid to ice transition case, and this is good because understanding this the phase transition is crucial, and poorly observed in the real atmosphere. Complimentary measurements at -40 degree C and -50 degree C are presented. It would be good to mention the difference between liquid origin and in situ cirrus, e.g. Kramer et al. A microphysics guide to cirrus clouds - Part 1: Cirrus types, ACP, 2016.

Added to section 2.2:

Continuous attempts to find an accurate cloud classification have led to suggestion of new definitions of liquid origin clouds and in situ cirrus (Krämer et al., 2016; Luebke et al., 2016). Liquid origin cirrus class comprises of ice crystals formed by homogeneous freezing of liquid drops farther below in the atmosphere which are uplifted into the cirrus temperature range. In situ cirrus class cloud may form in fast updraft triggered by jet streams or lee waves. This class has high IWC and many, small ice crystals. The formation mechanism in this case is insensitive to IN properties and dominated by homogeneous freezing. Our expansion profiles allow simulation of both types of clouds.

The two cases are not well described, neither is the motivation for doing the two types of expansion. The information for the deep convection (-30 degree C?) and in situ (-40 and -50 degree C) cases could be added to Table 1, and more discussion given to the differences. Crucially, how does supersaturation evolve over time in these simulations, especially with regard to liquid supersaturation in the -30 degree C case, and the icesubsaturation in all cases, for sublimation. A presentation of the cooling rate as well as the measured temperature would help here and additionally, what are the equivalent updraughts and are these reasonable.

- As stated in the text: This study tries to complement and extend the results previously obtained in the AIDA chamber with similar but not the same instruments (Järvinen et al., 2016c; Schnaiter et al., 2016) such as observation of morphological features together with an intercomparison of instrumental measurements of cloud particles.

- The motivation is the presence of spherical ice in different scenarios as mentioned in the "implications to atmospheric measurements" section, which is ambiguously detected by optical instruments. This is the reason for submission to AMT.
- The cooling rate and the initial temperature are presented in table 1, The SSice and SSwater plots were added. The expansion speed and the equivalent updraft can be estimated from the pressure drop curve. The equivalent updrafts presented in this paper are in the range of 4 – 11 m s^{-1}, which are slightly high, yet atmospherically relevant. Much higher updrafts were observed in the atmosphere by Yang et al. (2016); May and Rajopadhyaya, (1999); Zipser et al. (2006); Heymsfield et al. (2010); Hamada and Takayabu, (2016) and others. However, the vertical velocities are not the core of this asphericity detection and instrument intercomparison paper.
- Additional text was added about in situ and liquid origin clouds (see previous point).

The abstract claims to measure the response of four probes, but the reasons for the differences between the probes, e.g. technique, sample volumes, wavelengths, collection angles, are not explored in much detail.

I assume the referee suggests to explain the reasons we used the different probes and their unique abilities in the Methodology section, rather than explaining the differences between the instruments i.e. their technical specifications in much more details which are available elsewhere (e.g. Lawson et al., 2001, Nichman et al., 2016; Jarvinen et al., 2016b, Jarvinen et al., 2016a).

These different techniques are the key to understand the differences in observations. Each instrument is used to represent a single common optical discrimination method of optically spherical particles from asphericals. Each instrument has a separate description subsection in the text. Information about wavelengths, sample volume and collection angles are described in the text but some more details (e.g. sampling, wavelenghs, collection angles etc.) were added.

Also there is no information on how the airborne and PPD2K probes were aspirated, and what flow rates and particle rates were encountered. It would be good to provide information on the numbers of particles per second that the probes encountered so that coincidence can be ruled out, and a comparison against what they are designed for, when fitted to an aircraft. There is a brief discussion of coincidence, but no evidence that it was not present.

Answered in point 6 to Referee 1, the sampling air speed was approx.. 4 m s^{-1}
CASPOL measurements in ice clouds did not reach the coincidence threshold described by Lance et al., 2012.
In the reported CASPOL measurements, the highest particle rate was 500 # s^{-1}. The average rate was most of the time below 100 # s^{-1}. The maximum single particle detection of polarisation is 292 # s^{-1}.
PPD had a sample flow of 5 lpm and a sheath flow of 2 lpm. Count rates: <7000 s^{-1} (1276.05), <5500 s^{-1} (1291.07), <4000 s^{-1} (1291.12), <6000 s^{-1} (1291.01), <900 s^{-1} (1298.12).
Coincidence probability is shown in response to Referee 1 (page 18).
3V-CPI has low statistics rather than a coincidence problem.
For these reasons we rule out coincidence.

• page 4 line 9 - this subject is a good exploitation of the additional cooling rate available in this particular chamber
We agree.

• page 4 lines 11 to 14 - There seems to be a mismatch between consistent frozen mass (IWC) and smaller, more numerous particles, or higher IWC.

This is clearly an incorrect citation, thank you and Referee1 for bringing it to our attention.
Text amended:
"This is clearly shown by Ackerman et al. (2015) where ice particle mass distributions in homogeneous freezing for stronger updrafts produce substantially smaller ice particles".

• page 4 line 12 - Ackerman 2015 has now been moved from ACPD to ACP
Amended.

• page 5 line 7 - the CCN and cloud particle number data in Table 1 seem to disagree with the words, that all CCN are activated at low concentrations. What about high concentrations? And how is Table 1 ordered?

They don’t disagree.

- CCN concentration is #/cc measured with CPC, cloud particle concentration is dN/dlogD, but more important is that the cloud instruments are limited by their detection range and can miss the activated CCN smaller or larger outside the range. Especially at higher concentration when the size of activated aerosol is much smaller.
- There are losses on the chamber walls and sampling lines.
- See response to point 2 of Referee1 about activated fraction.
- Table 1 is ordered chronologically by run number order

• page 5 line 11 - Referring to the figure and the supplementary material would be helpful here. Including the supersaturation in the figures should also be done if this information is available.

amended

2.3 Section 3 - Results and Discussion

This section is a fairly complicated read, and is a mixture of instrument artefacts, experimental design, and results. Some parts may be better in the instrument section (methodology), and others in a separate experimental design followed by results section. For example discussion of coincidence errors (e.g. page 8 lines 13-16) and the 3V-CPI discussion in section 3.2.

Discussion of coincidence errors (page 8 lines 13-16) were moved to Section 2.3.1.

The author would like to accentuate the title of this instrumental intercomparison paper, submitted to AMT. We think that the aforementioned part in the Results and Discussion section are part of the discussion of the instrumental intercomparison, the limitations of the instruments, the selection of thresholds and the comparison of the analysis to other studies. This discussion which surrounds results presented in figures 1b, 5, 6a is not part of the Methodology. Moreover the general analysis technique is already briefly mentioned in the methodology.

It isn’t clear how it is known that the supercooled liquid regime only lasts a few seconds, and if the phase change and temperature changes happen uniformly throughout the chamber or not. Is time zero defined when water saturation is reached?

- Supersaturation plots were added.

- The course of the experiment (e.g. temperature, humidity, relative changes of measured size distributions) is already indicative for the ongoing microphysical processes and changes of cloud type (droplet activation and growth, freezing, drop evaporation, ice growth, sublimation…).
- The characterisation of chamber temperature is currently studied by Dias et al., 2016 (in preparation). The chamber has two fans and vertical and horizontal temperature measurements at different distances into the chamber volume (more details can be found in Duplissy et al., 2016). We assume homogeneous mixing as has the previously published work.
- Time zero defined with first detected cloud particles.

Do the observations confirm the pathway for quasi-spherical ice formation as claimed? The presentation of results here doesn't make this clear.

The paper is about instrument intercomparison and aspherical fraction measurements.
Page 9, Line 13 : "Therefore, the nearly spherical particles observed (4–10 min and from 19 min onwards) are spherical ice and not liquid water droplets. In atmospheric measurements, such an aspherical fraction would normally be converted into an ice fraction. In this cloud simulation, at the end of the sublimation period, both instruments misinterpret the total ice fraction as spherical-liquid by 60%".
Figs. 5, 6a show measurements of quasi-spherical particles.

What specifically does "complex particles" refer to on page 8 line 10.
It is mentioned earlier in the introduction Page 4, Line 15, and later Page 9 Line 38 to Page 10 Line 2. (Please see response to Referee1 on p.10).

Section 3.3 - The needs to be specific mention of which probes are being compared where. The CPI data as presented here look very variable, and it is difficult to infer trends in particle asphericity

There is a clear mention of every probe on each comparison plot. There is also a specific mention in the text of the probes compared. Text amended to include mention of CPI in Figs.

6b, 6c. We also smoothed the CPI data, reduced the size of the marker and changed the colour to magenta for clarity of the trends.

The analysis of the SIMONE and CASPOL data in figure 6c is possibly accurate, but very difficult to assess on the time series (page 9 line 21). The early period and late period look as though they might have different behaviour that a more detailed analysis would confirm or refute.

Text added: "Additional studies are required to confirm these trends."

It seems in figure 7a that the particles all freeze at a similar time, independent of size, which seems like an important observation that warrants a more detailed discussion regarding the implications for atmospheric clouds.

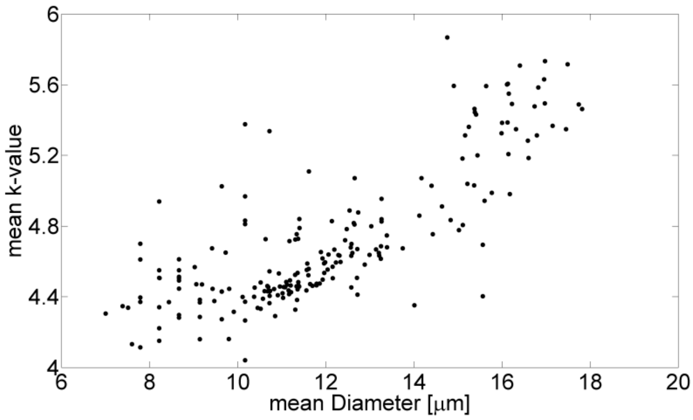
The complexity appears as the liquid droplets freeze, and it is valuable for the intercomparison, however, the threshold was not calibrated therefore further conclusions about stochastic and singular freezing could be too speculative in respect to atmospheric clouds.

There does not seem to be evidence presented here that the particles develop a frost layer - is this assumption just based on Jarvinen 2016c? Do they remain liquid in the centre for longer? And is droplet shattering during freezing important - is number constant?

- Yes, since we are complementing the previous set of experiments, a similar terminology was used.
- We did not have the instrumentation to analyse the phase of the core of the frozen droplets.
- The life time of the cloud is relatively short compared to the atmosphere, and reheating starts immediately at the end of the expansion. We did not observe splintering which normally produces high number of aspherical shapes which grow rapidly depleting any liquid water. However, detecting initial splinters at the point of production is extremely difficult.

I can’t see any size segregation in figure 7b that suggests the smaller particles are less complex - can some example scattering patterns illustrate this?

As the ice sublimates and decreases in size (Fig 7a), the Median K value (complexity) deceases (Fig 7b) – same time scale. Pattern D marked in Fig 7a illustrates the lower diameter sublimated quasi-spherical ice at this period of time (x axis). Mean complexity vs mean diameter shown in the figure below:



The limitations on CASPOL in phase determination are important and so this is a valuable observation - is it possible to put numbers on this, both size and concentration?

I don’t think it’s an explicit CASPOL limitation rather than a comparison limitation of two techniques. This limitation is not easily derived and is case dependent. It is apparent that one of the main limitations in asphericity measurements with CASPOL is the detection of only the first 292 particles s^{-1}. New versions of the instrument are now starting to address this limitation.

In cases where the size distribution is highly occupied by the smaller spherical particles and the concentration of the larger aspherical is lower, it is more probable that the single-particle 292 bins will be more occupied by the smaller particles in each second. It depends on sample size and whole population size, therefore the calculation of sampling margin of error is a key value for future comparisons.

This limitation is also demonstrated in Jarvinen et al., (2016a). (Please see answer to Referee1 on page 13).

The low occurrence or counts s^{-1} that we mention limit the fraction calculation, i.e. statistically meaningful fraction in every second. However it is not possible to provide a

single threshold for all cases, we can only estimate that occupation of more than 10% of polarisation bins would provide reasonable fraction accuracy.
More experiments need to be done to accurately define the threshold of size where the complexity is detected by scattered light polarisation.

Section 3.4 - The section comparing SIMONE and CASPOL is good for completeness of the study, but limited in impact. What are the effects of the difference in sample volume / path length, wavelengths, scattering angles and how does this impact the comparison?

All these differences impacting the scattering are obviously particle size and composition dependent and may intensify or decrease the scattering. The goal here is to show in which scenarios, despite the numerous differences, the polarisation ratio can be somewhat comparable for both instruments.

The should be more discussion on the implications of this comparison for real atmospheric measurements. There is no real quantitative analysis, and no limits or thresholds, in terms of size or concentration, that specify when a comparison might hold or fail.

Discussion of implications of this comparison for real atmospheric measurements would be incomplete without
1. The investigation of bigger particles which will affect the bulk measurement and won't be detected in the single particle CASPOL range (<50 micron).
2. Orientations effects e.g. in presence of high electric fields are also being studied as part of this campaign and seem to affect the comparison of single particle polarisation ratio vs bulk measurements. (e.g. Nichman et al., 2015)
Here we report the polarisation measurements of small sizes in several atmospheric scenarios; these findings can be used in future studies to explain discrepancies between single particle and bulk polarisation observations.
We have added thresholds in section 3.5

The correlations look weak in all cases. Would it help to average over a longer time period than 1 s?

The correlation doesn't have to be strong; we show instrument responses to different concentration and several sizes. There is a common trend to compare remote sensing and averaged path measurements to single particle polarisation and phase discrimination, however we can see in these examples that they are often incomparable.
Longer averaging in this case will make data more scattered due to the short time scale of expansion and low concentration.

Section 3.5 - This section should be expanded and more made of what the observations presented in the chamber mean for real experiments making measurements in the atmosphere, for example - what will happen at aircraft speeds to these techniques? It seems as though the main take-home messages of the work should be in this section.

Text added:
However, artefacts such as shattering, compression, and coincidence (Lance et al., 2012) that may occur in airborne measurement but do not occur in this setup may cause a mis-estimation of the ice fraction. In case small particle arrival rate and the concentration do not exceed the values reported here, the findings of this study may be applicable.

• page 12 line 3 - how high concentrations - which way does the error go, higher or lower fraction?

We did not observe coincidence issues in this study however we are aware they are ubiquitous in airborne measurements, therefore this sentence is simply a note to take into consideration. The error could go both ways: e.g. circular arrangment resulting from the alignment of several particles can be classified as spherical by 2DS or several coincident spheroids may be classified as a more complex shape. More often at higher concentration and coincidence, aspherical fraction will be overestimated.
Changed to: However, artefacts such as shattering, compression, and coincidence (Lance et al., 2012) that may occur in airborne measurement but do not occur in this setup may cause a misestimation of the ice fraction. In case small particle arrival rate and the concentration do not exceed the values reported here, the findings of this study may be applicable.

• page 12 line 6,7 - which techniques, single particle and ensemble measurements?

Changed to: Many single particle and ensemble measurements laboratory techniques in particular have proven difficult to adopt when translated to real atmospheric environments.

• page 12 line 8,9 - Is it possible to specify ranges of size and concentration, or thresholds where the two techniques are comparable

Changed to : Based on our analysis, ensemble depolarisation measurements of cloud particles at concentrations above 20 cm^{-3}, diameters below 15 μm and certain atmospheric conditions can be comparable to single particle airborne measurements.

2.4 Section 4 Conclusions

The conclusions as presented are useful, in that there is evidence presented that show the challenges to the atmospheric measurement community. However it would be good to see a quantitative assessment of when the probes can and can’t be used for what purpose.

For the purpose of aspherical discrimination in this paper we gave a quantitative assessment that unambiguous phase discrimination below 7microns is impossible with these probes. In the range 7-40 micron at certain conditions all the probes (used in this study) could misclassify the frozen droplets as liquid. The detection of complexity seems to have some advantage; however it is not clear how accurate it will be in random atmospheric environments.

Each of these instruments on its own is not ideal for small particle measurements, however a combined detector of all optical properties in the same sample volume measuring diffraction and polarisation at several adjacent wavelengths could provide the necessary resolution for more accurate phase discrimination.

Increasing resolution in future probes may help, but how much further given the optical limits.

There are now various techniques to push the optical limits further (e.g. Fung et al., 2009). In my opinion the technology is already available. The main concerns are price, safety, regulations and weight/size of the instrument.

Also, despite the limitations the current probes need to be exploited,
but there is no clear message on how to do this.

The instruments should be used according to the published recommendations, there is nothing wrong with them (besides the known problems), PSD can be measured without major problems within the instrument size range. However composition/asphericity comparisons such as these, between PPD and CASPOL or SID and CASPOL, should be done for narrow size distributions when all particle sizes are within the overlapped range of the instruments (before filtering). Otherwise a sampling error is introduced. Measured phase properties at concentration below coincidence thresholds and with high particle interarrival time would be more accurate. This can be problematic for mixed phase clouds.

Recommendations will be added:

While PSD measurements are reliable, phase discrimination should be verified by multiple instruments; therefore the vast majority of the measured particle sizes have to be in the overlapped detection range of CASPOL and other phase discriminating instruments. Future efforts should aim on calibration of scattering pattern analysis thresholds and interpretation of the reported complexity using offline high resolution microscopy. In airborne measurements, small particle detection is often contaminated by shattering and coincidence which should be addressed before any comparison can be made. It should be highlighted that the phase of small spherical particles with low complexity cannot be unambiguously defined with any of these instruments.

The conclusions refer to particles less than 60 microns, although CASPOL only measures up to 50 microns and particles in the study are smaller than this. It is not clear how these results will apply to other probes, especially impactors. Future probes may have better resolution detection, but depending on the size of surface complexity the limit could be the optical wavelengths used.

- Text was amended to 40 microns
- For quasi spherical frozen droplets as shown in Figure 6a, an impactor or replicator imprint will have the same difficulties to derive the phase of the particles once the ice has melted and spherical imprint remains. The subtle submicron features are harder to see on cured polymers due to small molecule leaching, polymer degradation, porosity or low compatibility of the sample with high resolution techniques.

In the case of online imaging (e.g. VIPS) the resolution doesn't allow accurate discrimination of the phase as described in this paper.

- Another option is a continuous scan over wavelengths to see how the scattering and polarisation are changing.

Typographical errors

• page 3 line 19 - 'a' climatic impact, or climatic importance?

amended

• page 3 line 24 - cases plural?

amended

• page 9 line 10 - ice fraction is referred to as aspherical fraction elsewhere

amended

• page 9 line 13 - is this from 19 min onwards?

amended

• page 9 line 31 - replace to with of

amended

• page 9 line 32 - with respect to

amended

• page 10 line 36 - start a new paragraph?

The whole paragraph describes small scale complexity vs. smooth particles, it looks more coherent as a single paragraph.

• page 10 line 28 - its, no apostrophe

amended

• page 10 line 33 – patterns

amended

• page 11 line 22 - averaging deviation

amended

• page 11 line 21 - capitalisation of lowest

amended

• page 11 line 37,38,39 - suggest rewrite for clarity

Changed to: In any case of small quasi-spherical particle detection at sub-zero temperatures in the atmosphere, we recommend to use multiple instruments for intercomparative analysis.

• page 11 line 27 - Implications "For"....

amended

• page 12 line 36 - is the comma required?

amended

Figure comments

• figure 3 - font size of axis titles

Font size corrected

• figure 6 - linear or circular?

Figs. 6a,c Linear, Fig. 6b Circular as stated

• figure 7b - the complexity line is very faint and hard to see

corrected

• figure 8 - difficult to read and extract the take home message, especially when there are lots of high concentrations data points. Font sizes are all different.

The concentration markers and font sizes have been corrected. Text added in the caption: “Better correlation is observed for clouds with higher concentration of small particles”.

References

Ackerman, A. S., Fridlind, A. M., Grandin, A., Dezitter, F., Weber, M., Strapp, J. W., and Korolev, A. V.: High ice water content at low radar reflectivity near deep convection – Part 2: Evaluation of microphysical pathways in updraft parcel simulations, Atmos. Chem. Phys., 15, 11729-11751, doi:10.5194/acp-15-11729-2015, 2015.

Baran, A. J. and Labonnote, L.-C.: A self-consistent scattering model for cirrus, I: The solar region, Q. J. Roy. Meteorol. Soc., 133, 1899–1912, 2007.

Baran, A. J.: From the single-scattering properties of ice crystals to climate prediction: a way forward, Atmos. Res., 112, 45–69, doi:10.1016/j.atmosres.2012.04.010, 2012.

Barrie, L. A.: Background pollution in the Arctic air mass and its relevance to North American acid rain studies, Water, Air, and Soil Pollution, 30, 765-777, 10.1007/bf00303343, 1986.

Buck, Buck Research CR-1A User's Manual, Appendix 1, 1996.

Connolly, P. J., Flynn, M. J., Ulanowski, Z., Choularton, T. W., Gallagher, M. W., and Bower, K. N.: Calibration of the Cloud Particle Imager Probes Using Calibration Beads and Ice Crystal Analogs: The Depth of Field, Journal of Atmospheric and Oceanic Technology, 24, 1860-1879, doi: 10.1175/JTECH2096.1, 2007.

Curry, J. A., Ebert, E. E. and Herman, G. F. (1988), Mean and turbulence structure of the summertime Arctic cloudy boundary layer. Q.J.R. Meteorol. Soc., 114: 715–746. doi:10.1002/qj.49711448109

de Boer, G., Shupe, M. D., Caldwell, P. M., Bauer, S. E., Persson, O., Boyle, J. S., Kelley, M., Klein, S. A., and Tjernström, M.: Near-surface meteorology during the Arctic Summer Cloud Ocean Study (ASCOS): evaluation of reanalyses and global climate models, Atmos. Chem. Phys., 14, 427-445, doi:10.5194/acp-14-427-2014, 2014.

De Mott, P.J.; Möhler, O.; Stetzer, O.; Vali, Gabor; Vali, G.; Levin, Z.; Petters, M.D.; Murakami, M.; Leisner, T.; Bundke, U.; Klein, H.; Kanji, Z.A.; Cotton, R.; Jones, Hazel; Benz, S.; Brinkmann, M.; Rzesanke, D.; Saathoff, H.; Nicolet, M.; Saito, A.; Nillius, B.; Bingemer, H.; Abbatt, J.; Ardon, K.; Ganor, E.; Georgakopoulos, D.G.; and Saunders, C. : Resurgence in Ice Nuclei Measurement Research, Bulletin of the American Meteorological Society, 92.12, 1623-1635, 2011.

Duplissy, J., Merikanto, J., Franchin, A., Tsagkogeorgas, G., Kangasluoma, J., Wimmer, D., Vuollekoski, H., Schobesberger, S., Lehtipalo, K., Flagan, R., Brus, D., Donahue, N., Vehkämäki, H., Almeida, J., Amorim, A., Barmet, P., Bianchi, F., Breitenlechner, M., Dunne, E., Guida, R., Henschel, H., Junninen, H., Kirkby, J., Kürten, A., Kupc, A., Määttänen, A., Makhmutov, V., Mathot, S., Nieminen, T., Onnela, A., Praplan, A., Riccobono, F., Rondo, L., Steiner, G., Tome, A., Walther, H., Baltensperger, U., Carslaw, K., Dommen, J., Hansel, A., Petäjä, T., Sipilä,M., Stratmann, F., Vrtala, A.,Wagner, P.,Worsnop, D., Curtius, J., and Kulmala,M.: Effect of ions on sulfuric acid-water binary particle formation II: Experimental data and comparison with QC-normalized classical nucleation theory, Journal of Geophysical Research: Atmospheres, doi:10.1002/2015JD023539, 2016.

Fung, R., Shneerson, V., Saldin, D. K., and Ourmazd, A.: Structure from fleeting illumination of faint spinning objects in flight, Nat Phys, 5, 64-67, http://www.nature.com/nphys/journal/v5/n1/suppinfo/nphys1129_S1.html, 2009.

Gayet, J.-F., Mioche, G., Shcherbakov, V., Gourbeyre, C., Busen, R., and Minikin, A.: Optical properties of pristine ice crystals in mid-latitude cirrus clouds: a case study during CIRCLE-2 experiment, Atmos. Chem. Phys., 11, 2537-2544, doi:10.5194/acp-11-2537-2011, 2011.

Hamada, A., and Takayabu, Y. N.: Convective cloud top vertical velocity estimated from geostationary satellite rapid-scan measurements, Geophysical Research Letters, 43, 5435-5441, 10.1002/2016GL068962, 2016.

Heymsfield, G. M., Tian, L., Heymsfield, A. J., Li, L., and Guimond, S.: Characteristics of Deep Tropical and Subtropical Convection from Nadir-Viewing High-Altitude Airborne Doppler Radar, J. Atmos. Sci., 67, 285–308, 2010.

Hoyle, C. R., Fuchs, C., Järvinen, E., Saathoff, H., Dias, A., El Haddad, I., Gysel, M., Coburn, S. C., Tröstl, J., Bernhammer, A.-K., Bianchi, F., Breitenlechner, M., Corbin, J. C., Craven, J., Donahue, N. M., Duplissy, J., Ehrhart, S., Frege, C., Gordon, H., Höppel, N., Heinritzi, M., Kristensen, T. B., Molteni, U., Nichman, L., Pinterich, T., Prévôt, A. S. H., Simon, M., Slowik, J. G., Steiner, G., Tomé, A., Vogel, A. L., Volkamer, R., Wagner, A. C., Wagner, R., Wexler, A. S., Williamson, C., Winkler, P. M., Yan, C., Amorim, A., Dommen, J., Curtius, J., Gallagher, M. W., Flagan, R. C., Hansel, A., Kirkby, J., Kulmala, M., Möhler, O., Stratmann, F., Worsnop, D. R., and Baltensperger, U.: Aqueous phase oxidation of sulphur dioxide by ozone in cloud droplets, Atmos. Chem. Phys., 16, 1693-1712, doi:10.5194/acp-16-1693-2016, 2016.

Jackson, R. C., G. M. McFarquhar, A. V. Korolev, M. E. Earle, P. S. K. Liu, R. P. Lawson, S. Brooks, M. Wolde, A. Laskin, and M. Freer (2012), The dependence of ice microphysics on aerosol concentration in arctic mixed-phase stratus clouds during ISDAC and M-PACE, J. Geophys. Res., 117, D15207, doi:10.1029/2012JD017668.

Järvinen, E., Schnaiter, M., Mioche, G., Jourdan, O., Shcherbakov, V. N., Costa, A., Afchine, A., Krämer, M., Heidelberg, F., Jurkat, T., Voigt, C., Schlager, H., Nichman, L., Gallagher, M., Hirst, E., Schmitt, C., Bansemer, A., Heymsfield, A., Lawson, P., Tricoli, U., Pfeilsticker, K., Vochezer, P., Möhler, O., and Leisner, T.: Quasi-Spherical Ice in Convective Clouds, Journal of the Atmospheric Sciences, 73, 3885-3910, doi:10.1175/JAS-D-15-0365.1, 2016a.

Järvinen, E., Kemppinen, O., Nousiainen, T., Kociok, T., Möhler, O., Leisner, T., and Schnaiter, M.: Laboratory investigations of mineral dust near-backscattering depolarization ratios, Journal of Quantitative Spectroscopy and Radiative Transfer, 178, 192-208, http://dx.doi.org/10.1016/j.jqsrt.2016.02.003, 2016b.

Järvinen, E., Ignatius, K., Nichman, L., Kristensen, T. B., Fuchs, C., Hoyle, C. R., Höppel, N., Corbin, J. C., Craven, J., Duplissy, J., Ehrhart, S., El Haddad, I., Frege, C., Gordon, H., Jokinen, T., Kallinger, P., Kirkby, J., Kiselev, A., Naumann, K.-H., Petäjä, T., Pinterich, T., Prevot, A. S. H., Saathoff, H., Schiebel, T., Sengupta, K., Simon, M., Slowik, J. G., Tröstl, J., Virtanen, A., Vochezer, P., Vogt, S., Wagner, A. C., Wagner, R., Williamson, C., Winkler, P. M., Yan, C., Baltensperger, U., Donahue, N. M., Flagan, R. C., Gallagher, M., Hansel, A., Kulmala, M., Stratmann, F., Worsnop, D. R., Möhler, O., Leisner, T., and Schnaiter, M.: Observation of viscosity transition in α-pinene secondary organic aerosol, Atmos. Chem. Phys., 16, 4423-4438, doi:10.5194/acp-16-4423-2016, 2016c.

Krämer, M., Rolf, C., Luebke, A., Afchine, A., Spelten, N., Costa, A., Meyer, J., Zöger, M., Smith, J., Herman, R. L., Buchholz, B., Ebert, V., Baumgardner, D., Borrmann, S., Klingebiel, M., and Avallone, L.: A microphysics guide to cirrus clouds – Part 1: Cirrus types, Atmos. Chem. Phys., 16, 3463-3483, doi:10.5194/acp-16-3463-2016, 2016.

Lance, S.: Coincidence Errors in a Cloud Droplet Probe (CDP) and a Cloud and Aerosol Spectrometer (CAS), and the Improved Performance of a Modified CDP, Journal of Atmospheric and Oceanic Technology, 29, 1532-1541, doi:10.1175/JTECH-D-11-00208.1, 2012.

Luebke, A. E., Afchine, A., Costa, A., Grooß, J.-U., Meyer, J., Rolf, C., Spelten, N., Avallone, L. M., Baumgardner, D., and Krämer, M.: The origin of midlatitude ice clouds and the resulting influence on their microphysical properties, Atmos. Chem. Phys., 16, 5793-5809, doi:10.5194/acp-16-5793-2016, 2016.

May, P.T., and Rajopadhyaya, D. K.: Vertical Velocity Characteristics of Deep Convection over Darwin, Australia, Mon. Weather, Rev., 127, 1056–1071, 1999.

McFarquhar, Greg M; Ghan, Steven; Verlinde, Johannes; Korolev, Alexei; Strapp, J Walter; et al. Bulletin of the American Meteorological Society92.2 (Feb 2011): 183-201.

Nichman, L., Järvinen, E., Wagner, R., Dorsey, J., Dias, A., Ehrhart, S., Kirkby, J., Gallagher, M.W., Saunders, C.P.: Ionization, Charging and Electric Field Effects on Cloud Particles in the CLOUD Experiment, Abstract AE31B-0442 presented at 2015 Fall Meeting, AGU, San Francisco, Calif., 14-18 Dec, 2015.

Nichman, L., Fuchs, C., Järvinen, E., Ignatius, K., Höppel, N. F., Dias, A., Heinritzi, M., Simon, M., Tröstl, J., Wagner, A. C., Wagner, R., Williamson, C., Yan, C., Connolly, P. J., Dorsey, J. R., Duplissy, J., Ehrhart, S., Frege, C., Gordon, H., Hoyle, C. R., Kristensen, T. B., Steiner, G., McPherson Donahue, N., Flagan, R., Gallagher, M. W., Kirkby, J., Möhler, O., Saathoff, H., Schnaiter, M., Stratmann, F., and Tomé, A.: Phase transition observations and discrimination of small cloud particles by light polarization in expansion chamber experiments, Atmos. Chem. Phys., 16, 3651-3664, doi:10.5194/acp-16-3651-2016, 2016.

Shcherbakov, V., Gayet, J.-F., Baker, B. and Lawson, P.: Light scattering by single natural ice crystals, J. Atmos Sci., 63(5), 1513– 1525, 2006.

Schnaiter, M., Järvinen, E., Vochezer, P., Abdelmonem, A., Wagner, R., Jourdan, O., Mioche, G., Shcherbakov, V. N., Schmitt, C. G., Tricoli, U., Ulanowski, Z., and Heymsfield, A. J.: Cloud chamber experiments on the origin of ice crystal complexity in cirrus clouds, Atmos. Chem. Phys., 16, 5091-5110, doi:10.5194/acp-16-5091-2016, 2016.

Vochezer, P., Järvinen, E., Wagner, R., Kupiszewski, P., Leisner, T., and Schnaiter, M.: In situ characterization of mixed phase clouds using the Small Ice Detector and the Particle Phase Discriminator, Atmos. Meas. Tech.,9, 159-177, doi: 10.5194/amt-9-159-2016, 2016.

Yang, J., Wang, Z., Heymsfield, A. J., and French, J. R.: Characteristics of vertical air motion in isolated convective clouds, Atmos. Chem. Phys., 16, 10159-10173, 10.5194/acp-16-10159-2016, 2016.

Zhang, D., Wang, Z., Heymsfield, A., Fan, J., and Luo, T.: Ice Concentration Retrieval in Stratiform Mixed-Phase Clouds Using Cloud Radar Reflectivity Measurements and 1D Ice Growth Model Simulations, Journal of the Atmospheric Sciences, 71, 3613-3635, 10.1175/jas-d-13-0354.1, 2014.

Zipser, E. J., Cecil, D. J., Liu, C., Nesbitt, S. W., and Yorty, D. P.: Where are the most intense thunderstorms on Earth?, B. Am. Meteorol. Soc., 87, 1057–1071, 2006.