

# ***Interactive comment on “Intercomparison study and optical asphericity measurements of small ice particles in the CERN CLOUD experiment” by Leonid Nichman et al.***

## **Anonymous Referee #3**

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## **1 Comments**

This study attempts to understand observations of small ice and liquid in simulated clouds at the CERN-CLOUD (European Organisation for Nuclear Research - Cosmics-Leaving-OUTdoor-Droplets ) chamber, using laboratory instruments PPD2K (Particle Phase Discriminator mark 2 - Karlsruhe) and SIMONE (Scattering Intensity Measurement for the Optical detectionN of icE) and airborne instruments CASPOL (Cloud and Aerosol Spectrometer with Polarisation) and 3V-CPI (3 View Cloud Particle Imager).

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

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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. 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.

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. 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.

## 2 Specific sections

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

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## 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. 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).

## 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.

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 ice-subsaturation 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.

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. 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.

- page 4 line 9 - this subject is a good exploitation of the additional cooling rate available in this particular chamber
- 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.
- page 4 line 12 - Ackerman 2015 has now been moved from ACPD to ACP
- 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?
- 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.

### 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.

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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? Do the observations confirm the pathway for quasi-spherical ice formation as claimed? The presentation of results here doesn't make this clear. What specifically does "complex particles" refer to on page 8 line 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.

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.

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. 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? 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?

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?

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? There should be more discussion on the implications of this comparison for

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

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.

- page 12 line 3 - how high concentrations - which way does the error go, higher or lower fraction?
- page 12 line 6,7 - which techniques, single particle and ensemble measurements?
- page 12 line 8,9 - Is it possible to specify ranges of size and concentration, or thresholds where the two techniques are comparable

## 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. Increasing resolution in future probes may help, but how much further given the optical limits. Also, despite the limitations the current probes need to be exploited, but there is no clear message on how to do this. 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

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depending on the size of surface complexity the limit could be the optical wavelengths used.

### 3 Typographical errors

- page 3 line 19 - 'a' climatic impact, or climatic importance?
- page 3 line 24 - cases plural?
- page 9 line 10 - ice fraction is referred to as aspherical fraction elsewhere
- page 9 line 13 - is this from 19 min onwards?
- page 9 line 31 - replace to with of
- page 9 line 32 - with respect to
- page 10 line 36 - start a new paragraph?
- page 10 line 28 - its, no apostrophe
- page 10 line 33 - patterns
- page 11 line 22 - averaging deviation
- page 11 line 21 - capitalisation of lowest
- page 11 line 37,38,39 - suggest rewrite for clarity
- page 11 line 27 - Implications "For"....
- page 12 line 36 - is the comma required?

## 4 Figure comments

- figure 3 - font size of axis titles
- figure 6 - linear or circular?
- figure 7b - the complexity line is very faint and hard to see
- 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.

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