Review to “Using depolarization to quantify ice nucleating particle concentrations: a new method” by Zenker et al. AMTD, 2017

The manuscript by Zenker and coworkers describes the application of a new method to discriminate particle types in continuous flow diffusion chamber (CFDC) studies using a depolarization signal obtained from the Aerosol Spectrometer with POLarization (CASPOL). The work is motivated by the difficulties faced when particle type discrimination is purely based on particle size (“traditional method”), as particles of the same size do not necessarily need to be of the same type. Using a set of training data, where only ice crystals, aerosol particles or (cloud) droplets exist the authors show how the CASPOL depolarization signal can be used to differentiate these particles types based on optical signatures. A linear regression fit model is then used to optimize the depolarization ratio value considered as (threshold) criterion to differentiate particle types, concluding with an optimal value of 0.3. The corresponding linear regression is then used to calculate the ice nucleating particle (INP) concentration for an extensive CFDC data set and the results for the “new” and the “traditional” method are compared.

I believe that the topic of reliable particle type discrimination in CFDC studies (when operating under water droplet breakthrough (WDBT) conditions) is inherently complex and needs to be addressed in the future. This manuscript certainly provides motivation to do so and the presented results show evidence that using depolarization ratio can contribute to a more accurate discrimination of particle type in CFDC studies than is currently done by size discrimination and ultimately leads to a better quantification of INP concentrations.

However, the manuscript at the current state would benefit from restructuring and major revisions to clarify certain key aspects of the data analysis and interpretation. Once all concerns given in the following are properly addressed, this manuscript may be suitable for publication in AMT.

General Comments:

Section 1: Please focus more on the core topic of the manuscript and provide background for the particle discrimination in CFDC studies. I also encourage the authors to motivate the need for a better particle type/phase discrimination in order to more clearly indicate the additional value obtained from new methods as presented in this manuscript.

Section 2: This section requires restructuring and currently misses important technical details for the instruments used (e.g. TAMU CFDC) or appropriate references. Please add more instrumental details to the manuscript.

In Section 3, the creation of the simulated data sets and implementation of the regression model remains unclear to me. I struggled to follow how the optimal depolarization ratio threshold is identical to the one presented in Section 3.3, which I assumed at this stage to be empirical. The justification on using a linear regression model and the implicated assumptions on the data is entirely missing and only legitimated by indicating that other work has used linear regression models. Please add the justification for doing so. Also expand on how the choice of another depolarization ratio threshold does influence your results. Lastly, the comparison of the TAMU data to the CSU data stays unclear. As presented in the current manuscript the usage of different cut sizes due to instrumental differences is irritating and needs clarification. A quantification of the (range) extension for the operating conditions of the TAMU CFDC when applying the new method along with the associated error should be included.

Specific comments:

- P. 1-3, *Introduction*: The authors state the goal of the presented paper to be the development of a new method to quantify INP through a more reliable (phase) discrimination of particles exiting a CFDC, especially when operated under WDBT conditions (cf. p. 1, l. 15-18, p. 4, l. 13-19).

In the introduction, the authors carefully describe the importance of ice and mixed-phase clouds and go on to discuss different ice nucleation pathways and INP characteristics (p. 2, l. 3- 13 and p. 2, l. 19-20). After a brief discussion about the hydrometeor discrimination by LIDAR measurements using depolarization signals (p. 2 l. 25 – p. 3 l. 8) the authors give a detailed overview of the CFDC history and the improvements done to CFDCs (p. 3 l. 16-25). None of the topics mentioned above adds significant information to the topic discussed in the article, namely the correct discrimination of cloud particle type (phase).

However, the introduction misses a clear description of the current limitations of particle phase discrimination in CFDC studies as well as a motivation how such limitations affect past and current INP measurements, using CFDCs. P. 4 l. 4-13 give details about how other studies differentiate particle phase, without discussion of the general limitations.

Without this discussion it becomes very hard for the reader to correctly judge the quality of available CFDC data and recognize the need for development of new instrumentation to improve discrimination of hydrometeor type. I suggest to add some references here as well. Finally, the benefit of new methods, as described in the presented study becomes clearer.

I recommend major changes to the introduction of the presented paper by considerable shorten or remove some of the topics mentioned above and focusing on background needed to understand the (size dependent) discrimination of particle phase and associated limitations, to better put the current study into context.

- P. 1, l. 12: Please change “observed” to “measured”.
- P. 1, l. 15: Please change for clarification: “…under which discrimination of hydrometeor phase and thus determination of INP concentrations based on hydrometeor size fails.”
- P. 1, l. 18-19: Please clarify this statement. It is not a challenge of WDBT that needs to be overcome, as WDBT forms an integral component of any CFDC study if operated at given conditions, but rather the challenge to reliable discriminate particle phase of the particles exiting a CFDC once WDBT conditions are met.
- P. 1, l. 25: Please change “complicated” to “complex”.
- P. 1, l. 26: Please clarify whether “precipitation” refers to spatial/temporal distribution of precipitation, precipitation formation or precipitation in general.
- P. 2, l. 2: Leave out “our”.
- P. 2, l. 8: Leave out “becomes”
- P. 2, l. 11: Please insert: “aerosol particle…”
- P. 2, l. 12: Please change to: “aerosol particle collides with a supercooled water droplet and …”
- P. 2, l. 20 : Delete “Field”
- P. 2 , l.23: Delete “other”
- P. 2, l. 28: Delete “can”
- P. 3, l. 4: Please change to: “…components of the LIDAR signal retrieved from…”
- P. 3, l. 13-15: The first argument only applies to field measurements, when CFDCs are used to characterize ambient INP concentrations. However, the data you present here result from laboratory measurements, where the number of aerosol particles entering the cloud chamber (and thus the number of INPs) can be varied by the experimentalist, making this argument irrelevant for this study. Please revise this section by making it clearer, that this is particularly a limitation of CFDC field studies.
- P. 3, l. 10-22: Shorten this paragraph and to keep the focus on the topic of your manuscript.
- P. 3, l. 23: Please change to “… (CLIMET Inc., Model No. CI-3100) …”
- P. 4, l. 13: Delete “to detect INP”
- P. 5, l. 4: Please change to: “… are generated, suspended in dry synthetic air…”

- P. 5, l. 7: Please specify whether aerosol particles or populations of ice crystals and cloud droplets have been sampled from AIDA.
- P. 5, l. 9: Please add: "… of the TAMU CFDC-CASPOL measurements …"
- P. 5, l. 25: Please change "limited" to "small".
- P. 6, l. 1-4: Specify how *ice* saturation is maintained in the evaporation section of the CFDC given that you have hydrophobic Teflon walls.
  How much are the ice crystals evaporated when passing through the lower most 25 cm of the chamber? Can you show that the ice crystals remain in the sample flow?
- P. 6, l. 4: Please specify why the droplets in some cases only partially evaporate. The evaporation efficiency is a function of particle residence time in the evaporation section. Your description of TAMU is missing a statement about the flows and thus residence times used within the TAMU CFDC. Such a discussion is only very briefly given on p. 7, l. 7-8 and should be moved to the description of the TAMU operation.
  Details of the residence time are also required to understand how the authors are able to grow ice crystals as large as 40 µm in the CFDC, as suggested by Fig. 6.
- P. 6, l. 8-15: This description of cloud chamber preparation does not add to the topic discussed in the presented paper and should be moved to a supplement.
- P. 6, l. 17: Please change to: "… (CLIMET Inc., Model No. CI-3100)…"
- P. 6, l. 26: Please change to: "…backward scatter detector…"
- P. 6, l. 5: The position of the mass flow controllers should be specified. I assume these are located downstream of CASPOL?
- P. 7, l. 13: Please change to: "Temperature, …"
- P. 7, l. 17: Please change "ahead" to "upstream"
- P. 7, l. 18-19: Please specify how the background (BG) signal from the CFDC is taken into account in more detail. Given that the supersaturation at the position of the aerosol lamina is different before and after a RH scan, the background signal is likely to change from before to after the measurement. The statement between lines 16-19 suggest that there is not always a BG measurement before and after each RH scan ("and/or after"). This makes it hard to follow what BG signal is subtracted from your CFDC-CASPOL measurements.
- P. 7, l. 20-24: This sentence is misleading. I assume you refer to the usage of the optical particle counter and the associated size cut-off used to discriminate between ice crystals and cloud droplets when using the term "traditional analysis". This is in contrast to the p. 6, l. 16-19, where it is described that TAMU had been used with both, OPC and CASPOL, so in principle both detectors can be interpreted as the traditional detector technique/analysis method. I suggest to make a clearer distinction between these two cases (OPC vs. CASPOL as detector) and give a clear statement earlier in the manuscript what the "traditional analysis" refers to.
- P. 7, l. 27: This statement is misleading. There are no limitations of the OPC technique (discrimination purely based on size) discussed in Section 2.3. Please delete the part in brackets. The authors start a superficial discussion of the limitations by using and OPC and a size threshold to discriminate the phase of cloud hydrometeors at various points of their manuscript, e.g. p. 3. l. 15-16, p. 3, l. 23-25. However, a clear statement that under certain thermodynamic conditions within the TAMU CFDC, cloud droplets and ice crystals of the same size can be present, thus biasing a pure phase discrimination based on particle size, is missing. This should be discussed in the introduction.
- P. 8, l. 3: The authors mention the limitations of traditional methods, but do not discuss differences how the ice crystal size threshold may be chosen. Please give more details.
- P. 8, l. 8: What do the authors mean by positive or negative artifacts?
- P. 8, l. 12: The challenges are not really presented by WDBT, but are rather inherent to any optical method that uses size as a means of phase discrimination.

- P. 8, l. 14-21: This section is partly a repetition of the statement made on p. 5, l. 23. Besides, it should be clear to any reader that a particle of any type (aerosol, cloud droplet) larger than the cut size will be misclassified as ice crystal by the OPC when using size thresholds to define an ice phase.
- P. 8, l. 19-21: Are the authors trying to say that large aerosols are not counted as ice crystals in their detector and they can be distinguished from an ice crystal of the same size?
- P. 8, l. 16: The expression "higher supercooled temperatures" is not clear. The authors should indicate more clearly what they compare to and point out reasons why the new analysis method is particularly powerful at higher T. The only indirect hint for this is given by the statement in brackets on p. 8, l. 10.
- I recommend moving the discussion of section 2.4 to the introduction to motivate the development of the new method.
- P. 8, l. 25: Please be more general in a first statement. The goal, as far as I understand from the presented study, is to first distinguish more accurately between aerosol particles, ice crystals and cloud droplets and then in a second step quantify the INP, as you clearly write e.g. p. 4, l. 13.
- P. 9, l. 1: I suggest repeating the meaning of the different parameters again. E.g. "Similar to eq. (1) $B_{\perp,CAS}$ and $B_{||,CAS}$ denote the perpendicular and parallel components of the backscattering signal, respectively, and the subscript CAS refers to the CASPOL signal…."
- P. 8, l. 24: Section 2.5 describes CASPOL instrumental details and should be moved to the description of CASPOL in section 2.2.
- P. 9, l. 21: Please explain why the neutralizer prevents particle loss.
- P. 9, l. 22: Please change to: "the" before CASPOL
- P. 10, l. 15: I assume you are referring to CFDC-CASPOL measurements, right? Please clarify.
- P. 10, l. 20: Please clarify the source of your temperature uncertainty here. How can the temperature uncertainty here be much lower than the value given on p. 7, l. 10?
- P. 10, l. 15: Please change SS to saturation ratio formulation throughout the manuscript. This will avoid confusion as of the negative sign and make your figures more easily readable.
- P. 10, l. 20: Do you suggest that particles smaller than 2 µm are not necessarily frozen?
- P. 10, l. 23: Insert comma after "datasets"
- P. 11, l. 4-5: Please clarify the usage of optical signatures by Hu et al. (2009) and how this relates to your study.
- P. 11, l. 6: Delete "training"
- P. 11, l. 13: Please clarify whether D_p refers to the optical diameter measured with CASPOL or to another diameter measured with another device.
- P. 11, l. 17: Please extend your interpretation of why almost only ice crystals show high values for $B_{\perp}/F$ and what that implies.
- P. 11, l. 23: Insert point after "et al."
- P. 11, l. 24 : Please clarify why this is an *empirical* tool and how this affects the application to your data.
- P. 12, l. 4: "It is assumed that the CASPOL emits an incident beam that propagates along the z…"
  - Why is it only assumed? Can you verify this experimentally?
  - Which direction is the z-direction? A schematic figure defining the different parts of CASPOL along with a coordinate system will definitely improve your description here. Please add a figure to your supplement.
  - It is not the CASPOL, but the laser diode of the CASPOL that emits the light.
- P. 12, l. 8: Please change to: "…line linking particle (position) and detection point."
- P. 12, l. 12: Please insert commas: "… ratio, δ_{Model}, can …"

- P. 12, l. 15-16: Please insert commas: "…matrix, P_{ij}, the amplitude matrix, S_{ij}, and the scattering cross section, C_{sca}, …"
- P. 13, l. 13: Please replace "vs." by "as a function of"
- P. 13, l. 17: Please delete "the" in front of optical signatures.
- P. 13, l. 13-21: Please elaborate this discussion and give more details:
    - Below approx. 2 µm no modelled depolarization ratios are given for any of the ice crystals, making a comparison between aerosol particles and ice crystals as suggested in the text difficult (l. 18-19)
    - The authors discuss the differences in depolarization ratio as a function of ice crystal habit in the range 2-4 µm. However, there is a clear distinction also above 10 µm for e.g. hexagonal plates and hexagonal columns. This needs to be explained.
    - What are the uncertainties associated with the modelled results. Errors bars should be included for the individual data points to render a comparison possible at all.
- P. 14, l. 1: Please insert: "aerosol particles"
- P. 14, l. 2: Please insert: "… shown in Fig.1 …"
- P. 14, l. 4: Please change to: "Each nominal droplet size produced by the VOAG is treated as a separate population in the training data set and …"
- P. 14, l. 5: Please change to: "… in Fig. 1a …"
- P. 14, l. 10: Please clarify how the selection criterion for ice crystals (depolarization ratio > 0.3) is derived and how it is connected to the values discussed in Fig. 1 (cf. p. 11, l. 15).
- P. 14, l. 15-16: Why do the aerosol particles in Fig. 1c show a mode only in the constrained size range between 5 to 10 µm and not above 5 µm in general?
- P. 15, l. 12: Please specify what you mean by "the size mode". I think you are referring to the smaller mode of the bimodal size distribution described above.
- P. 15, l. 13-19: In section 3.3 you discuss the usage of a depolarization threshold of 0.3 to discriminate between different particle types ("nominal selection criteria for depolarizing ice crystals"). In Fig. 4b all of your particles have significantly lower depolarization values, even at times when you are supersaturated. Please clearly state, that water droplets cannot be present during the time period before 11:55 due to the fact of being subsaturated, to avoid any confusion with your threshold of 0.3 discussed earlier.
- P. 15, l. 16-18: This is not correct. It is not the mean depolarization ratio, which has a strong dependence on whether WDBT is occurring in the CFDC, or not. Analyzing the depolarization ratio, you can observe the moment when WDBT occurs in the CFDC. Please phrase that more carefully.
- P. 16, l. 17: The statement "… at colder temperatures of these runs" is misleading, as the center temperature in your CFDC stays constant for each of the two runs. Furthr, the two Snomax® cases presented are not labeled in the figure, such that the reader cannot assign a CFDC center temperature difference between the runs from the lines in Fig. 5.
- P. 16, l. 26-27: How does the error shown for the observed values compare to the instrumental uncertainty from CASPOL to determine the right depolarization ratio? Please add errorbars associated with the modelled results. Consider using standard error of the mean for normalization to number of observed particles at the different sizes.
- P. 16, l. 28: Please add for clarification: "… from all FIN-02 experiments and not only the Snomax® experiments discussed in Section 3.5."
- P. 17, l. 1: Remove "u" after 2.
- P. 17, l. 1-4: Consider deletion as you already reference to the description given in section 3.5.
- P. 17, l. 5-10: Do the authors have any idea what type of ice crystal is formed in the CFDC? Is this dependent on the aerosol type/experiment? Which of the modeled ice crystals is closest to the "CFDC ice crystals"?

- P. 17, l. 14: Please replace “region” by “population”.
- P. 17, l. 19: Why do the “CFDC ice crystals” show depolarization ratios < 0.35 for all sizes shown? It is unclear to me how this relates to the data shown in Fig. 5, where the majority of the ice crystals show larger depolarization ratios. In addition, none of your “CFDC ice crystals” would meet the 0.3 threshold in depolarization ratio discussed on p. 14. Please explain. Is this due to averaging over all FIN-02 experiments?
- P. 17, l. 23-26: More details about the “underestimation of the depolarization by CASPOL and the detection limit” along with an appropriate reference should be given. Should the underestimation of depolarization by CASPOL not preliminary affect smaller sized particles (that scatter in relatively less light)? Thus the discrepancies between modelled and observed results should decrease as a function of size, as the detection by CASPOL becomes more reliable?
- P. 18, l. 22-24: How does this statement fit to your data shown in Fig. 6 (cf. “CFDC ice crystals”)?
- P. 18, l. 25: This is contradictory to the values you state on p. 16, l. 8-9. Please clarify.
- P. 18, l. 27: Please clarify what signal to noise ratio you refer to.
- P. 19, l. 7: I suggest giving more details here, as referring to an “optimal threshold” at this point is confusing. This threshold comes out of your training data sets (Fig. 3). However, in Fig. 6 you show that application of this threshold is not sufficient to discriminate droplets and ice crystals for WDBT conditions anymore. There, using the term “optimal threshold” should be avoided.
- P. 19, l. 24-28: Please specify why the linear fit was done for the case of M = 1. It is not clear, why the fit derived from the M = 1 case, is applied to all the other data sets M = 2 to M = 50.
- P. 20, l. 4: Please replace “The Fig.” by “It”.
- P. 20 l. 9: Given that you describe an optimization problem, there should be one optimum and a range of acceptable values. Please justify your statement on p. 9, l.2
- P. 20, l. 5-8: You describe a threshold used to distinguish between ice crystals, droplets and aerosols, to then derive Eq. (9), which yields the number of ice nucleating particles. However, the number of ice crystals is usually way larger than the number of INP. Please explain in more detail, how you derive a “parametrization” for INP at this stage.
- P. 20, l. 22: Please add: “Each relative humidity scan…”
- P. 20, l. 23: Please replace: “Supersaturation” by “Saturation ratio”
- P. 20, l. 25: Before this statement the meaning of the circles and the asterisks needs to be introduced in the text.
- P. 21, l. 11: Please specify what the value of the CASPOL uncertainty refers to. Is this the depolarization ratio signal?
- P. 21, l. 23: Please quantify the detection limit of CASPOL or give an appropriate reference.
- P. 21, l. 22: Please replace “polluting” to “biasing”
- P. 22, l. 2: Please add: “...mean percent error (MPE)…”
- P. 22, l. 11-12: Please quantify the concentration rate, where the new method is applicable rather than stating “high concentrations” and quantify the “accuracy” indicated.
- P. 22, l. 16: The benefit from this last paragraph and the additional comparison to the CSU CFDC, along with different cut-sizes shown in Fig. 11, does not become clear. Please explain in more detail.
- P. 22, l. 16 – P. 23, l. 14: Is your new method not applicable to other CFDCS operated along with CASPOL at all?

Figures:

Figure 1:

- Please locate the axis ticks also outside of the subpanel boxes to increase readability.
- Please be consistent with the terminology defined in Eq. (2) and include the subscript "$_{CAS}$" in the axis labels (also on the y-axis).
- "$_{CAS}$" subscript should be included in terminology used in figure caption.
- Subpanels (a/d), (b/e) and (c/f) are plotted for the same datasets. However, the colorbars for the upper row of subpanels and the lower row of subpanels use different colorcoding, which renders a comparison difficult. I suggest to change this using the same range for the color scale.

Figure 2:

- X-axis (labels) should be read as log-scale.
- Please include model calculations for larger aerosol sizes, such that there is a size overlap for the different particle types. This is needed to justify your statement on p. 13, l. 18.
- Please delete the term "Model" in your legend, as this is redundant information from the y-axis label and the figure caption.
- Caption: Please insert comma after droplets.

Figure 3:

- Please add symbol for depolarization ratio in x-axis label of panel (b), for consistency.
- These are all size distributions measured with CASPOL, right? Was there any additional instrument used, e.g. an Aerodynamic Particle Sizer, to verify the size of the produced particles? If so, please add these information and graphs to a supplement.

Figure 4:

- What are these large particles prior to 10:45 CET? The authors mention (p. 5, l. 24) that no impactor was used during the FIN-02 campaign and that the number of large particles was limited. I suggest to add a number size distribution of the Snomax® sample shown in Fig. 4 to the appendix for clarification. How do these large particles in the range 5- 10 µm influence the depolarization ratio shown in Fig. 4b (see also your Fig. 3)? Please add a description to the discussion in the manuscript.
- I suggest showing Panels (a) and (b) as a function of saturation ratio w.r.t. water instead of time. Saturation ratio w.r.t. ice can then be given as a second/top x-axis for instance. There is no additional information given by time. By using saturation ratio w.r.t. water it will be easier for the reader to put the discussed WDBT into context. Indicating ice saturation ratio will help to identify the formation of ice crystals. The text on p. 15 should be changed accordingly and can make more clear what is meant with "normal operating conditions" (p. 15, l.8). Further, labels for "normal" and "WDBT" conditions in Fig. 4 could help.
- Please make axis ticks more visible (e.g. reduce thickness of axes) and add ticks to x-axes in Fig 4a/b.
- Add explanation for the horizontal dashed lines in the figure caption (see p. 7, l.20).
- Caption for panel c should include the case number and a reference to Table 1.

Figure 6:

- I suggest using log-scale for the x-axis.
- Even though you state that the error bars show standard deviations from the mean, they seem to be on the same order of magnitude. Please add error bars (e.g. for some of the data)

- Please change the label of the y-axis as the data is a mixture of modelled and observed depolarization ratios.

Figure 10:

- The x-axis label should read “traditional concentration”.

References:

Hu, Y., Winker, D., Vaughan, M., Lin, B., Omar, A., Trepte, C., Flittner, D., Yang, P., Nasiri, S. L., and Baum, B.: CALIPSO/CALIOP cloud phase discrimination algorithm, Journal of Atmospheric and Oceanic Technology, 26(11), 2293-2309, 2009