Authors' response to Anonymous Referee #4 comments on "Using depolarization to quantify ice nucleating particle concentrations: a new method" *by* Jake Zenker et al.

The authors thank the anonymous reviewer for the detailed comments, published on Aug 9. 2017. In the response below, we address each of the suggestions of the reviews.

Anonymous Referee #4 Received and published: 9 August 2017

This manuscript (Using depolarization to quantify ice nucleating particle concentrations: a new method by Zenker et al.) capitalizes on the ability of the CASPOL detection method to capture the depolarization information from particles, droplets and ice particles in the TAMU CFDC and identify them under different operating conditions. The method may be applicable to other systems but each CFDC is unique. The manuscript includes the development of a new empirical analysis method, to quantify ice nucleating particle concentrations and presents a way to deal with especially the data obtained during water droplet breakthrough, which is difficult to interpret. I believe that the manuscript topic does fall into the scope of AMT. Generally, the paper is readable, the analysis is carefully done and discussion points seem to be well supported by data. There is a limitation for this method in that higher concentrations only obtainable in a laboratory are applicable; the authors are upfront about this limitation.

Referee Comment: There are some major considerations that, if addressed, could strengthen the paper: The authors may want to consider strengthening the end of their introduction to describe in more details the trajectory of work presented in the paper. Such a road map is limited here and more details could be helpful. In the body, there is little text regarding the comparison but there is a lot of text with many details regarding the development of the empirical analysis, yet these seem equally weighted in the introduction.
Authors' response: As per this Referee's comment as well as those of another, the introduction has been significantly restructured, including a "road map" in the final paragraph of the section, as suggested here.
Authors' changes in the manuscript. Please see the new introduction as a whole.

Referee Comment: In terms of the training data, the text notes that no droplets below 2 µm were studied and this is reflected in figure 3. However, figure 6 shows training droplets at 0.7 µm. This is confusing. Further, since this size is a cut-off point for the analysis, it might be helpful to include smaller particles generation or to explain how the data in figure 6 was observed.
Authors' response: I would ask the referee to revisit the figure. No training droplets below 1 micron are plotted. The training droplets are cut at 1 micron to eliminate residual 2-propanol droplets that form in the generation of the olive oil droplets.
Authors' changes in the manuscript: To avoid confusion, Figure 6 has now been revised and does not show training droplets below 1 micron. The elimination of <1 micron diameter residual 2-propanol droplets is also stated in the text.

Referee Comment: There may be minor scientific issues associated with the depolarization theory (that section of the paper was difficult to follow and there seemed to me to be some confusion or missing information associated with representations of matrices, matrix elements and values and/or units). In particular, the section on page 12 surrounding equations 6-7 is especially confusing. The authors note that these equations deal with the amplitude matrix, but then their inclusion in the equation appears to be an element with only one index. Further, it would be helpful to explain this part of the model further. What do these relationships (eqn 6-7) represent? I see how they combine to create eqn 8 but why?
Authors' response: The text has been revised to indicate that not only one index is included. Also, equation 8 is required in the form presented here for direct comparison to the CASPOL which detects light over single band of back scattering angles. 168^o to 176^o. This was mentioned in the experimental section, but we now include it here as well.

Authors' changes in the manuscript: The text on page 13 now reads, "Using the following relations between the elements of scattering phase matrix, P_{ij}(i,j=1,2,3,4), and the elements of amplitude matrix, S_i (i=1,2,3,4), below,

$$|S_4(\theta)|^2 + |S_2(\theta)|^2 = (P_{11}(\theta) + P_{12}(\theta)) \times C_{sca}, \quad (6)$$

$$|S_4(\theta)|^2 - |S_2(\theta)|^2 = (P_{21}(\theta) + P_{22}(\theta)) \times C_{sca}, \quad (7)$$

where C_{sca} is the scattering cross-section of a particle. As described above, the CASPOL detects light over single band of back scattering angles. 168^o to 176^o. To compare to the CASPOL measurements, we define the mean modeled depolarization ratio over the angular range of 168° to 176° and is expressed below in Eq. (8).

$$\bar{\delta}_{Model}(168°:176°) = \frac{\int_{168°}^{176°}(P_{11}(\theta) + P_{12}(\theta) - P_{21}(\theta) - P_{22}(\theta))\ \sin(\theta)\,d\theta}{2\int_{168°}^{176°}(P_{11}(\theta) + P_{12}(\theta))\ \sin(\theta)\,d\theta} \quad (8)"$$

Referee Comment:
It would also potentially be helpful for the authors to further discuss the use of the T matrix model for dust (and ice)? A recent technical note (Koepke et al., ACP, 2015, 5947) may be helpful. Generally, the paper would be enhanced with some additional details, clarity or references (and/or possibly even information in the experimental section) associated with the model calculations.
Authors' response: To clarify, the ice crystal calculations were performed using improved geometric optics methods, while the dust calculations were performed using t-matrix. We have now added additional details regarding each of these methods, and have added more additional references.
Authors' changes in the manuscript: The text on pg. 14 ln 4-13 now reads, "To compute the scattering phase matrices of these models with specific sizes at CASPOL wavelength, we apply so-called improved geometric optics method (IGOM) for particle with relatively large size and the invariant imbedding T-matrix method (II-TM) for particles with relatively small sizes (Yang

et al., 1996; Bi et al., 2013; Bi and Yang, 2014; Johnson, 1988). The combination of these two methods is chosen because of the different size parameters of the aerosol and ice crystal populations. The T-matrix method is a highly accurate method for calculating scattering properties of atmospheric particles (Koepke et al., 2015; Brooks et al., 2004). However, it becomes impractical for large particles due to its excessive demands on the computational power. In contrast, the IGOM is accurate over the range of particle sizes over which the particle size to be much larger than the incident wavelength (Xu et al, 2017)."

Added references:

Brooks, S. D., O. B. Toon, M. A. Tolbert, D. Baumgardner, B. W. Gandrud, E. V. Browell, H. Flentje, and J. C. Wilson (2004), Polar stratospheric clouds during SOLVE/THESEO: Comparison of lidar observations with in situ measurements, *Journal of Geophysical Research-Atmospheres*, *109*(D2).10.1029/2003jd003463.

Koepke, P., J. Gasteiger, and M. Hess (2015), Technical Note: Optical properties of desert aerosol with non-spherical mineral particles: data incorporated to OPAC, *Atmospheric Chemistry and Physics*, *15*(10), 5947-5956.10.5194/acp-15-5947-2015.

Xu, G., B. Sun, S. D. Brooks, P. Yang, G. W. Kattawar, and X. Zhang (2017), Modeling the inherent optical properties of aquatic particles using an irregular hexahedral ensemble, *Journal of Quantitative Spectroscopy & Radiative Transfer*, *191*, 30-39.10.1016/j.jqsrt.2017.01.020.

Referee Comment:
Overall, there is a lack of consistency within the text and figures where attention to detail would help. This is true, especially with the ordering of the types of particles within the different sections and also within the figures and captions. Further axis labels should include units where possible. A specific example is that in Fig. 6, there are both model and experimental results displayed but the y-axis includes the model label and the x-axis is missing units. Some additional specifics are included below.
Authors' response: Thank you for addressing these specific inconsistencies. We have edited many of the figures in response to this comment and others.
Authors' changes in the manuscript: Please see the manuscript for revised figures. Labels and captions are now be consistent.

Specific comments:
Referee Comment: Pg 9, line 24: e is missing from the
Authors' response: Added.

Referee Comment: Pg 10, line 11: “both” is unnecessary and confusing.
Authors' response: Deleted.

Referee Comment: Pg 10, line 19-20: In final copy, watch for placement of the minus sign

Pg 11, line 6-7: placement of training
Authors' response: This has now been fixed.

Referee Comment: Pg 11, line 8: based on the figure, the authors mean total backscatter vs. depolarization ratio. I'd also suggest reversing the order in the follow up sentence on lines 8-10.
Authors' response: The text in this section (now pg 12, ln 14-17) has been revised has been revised for clarity.

Referee Comment: Pg 12, line 7: k is in eqn 3, but omega and t are not present. Is the equation missing time dependence? Also, r is not defined until line 11. Pg 12, equation 4: related to above, are both matrices and matrix elements included? Pg 12, missing comma after Pij
Authors' response:
Yes, indeed Equation, 3 should be written as

$$\boldsymbol{E}_i = \begin{pmatrix} E_{\parallel i} \\ E_{\perp i} \end{pmatrix} e^{i(kz-\omega t)} = \begin{pmatrix} E_{\parallel i} \\ 0 \end{pmatrix} e^{i(kz-\omega t)},$$

but note that $e^{i(\omega t)}$ is later omitted when express the relation between incident and scattered field in Eq(4) , because the scattering is assumed to be elastic. As in the original, r is introduced when it first used.
Authors' changes in manuscript. Equation 3 has been corrected and "…scattering is assumed to be elastic." is now included. The elements of the amplitude matrix in Equation 4 are now defined: S_i (i=1,2,3,4) in Eq(4). Also, the text as been added: "Note that $e^{i(\omega t)}$ term is omitted since the scattering is assumed to be elastic."

Referee Comment: Pg 12, missing comma after Pij
Authors' response: Added. Thank you.

Referee Comment: Pg 14, line 5: I think you mean Fig. 3a here.
Authors' response: Corrected. Thank you.

Referee Comment: Pg 14, lines 14-17: This is confusing, please clarify. In figure 3b, it seems the % of total population of all particles having a depolarization ratio of 0.2 is close to 100%. How do other ratios exist for the population? This also makes interpretation of values in the text confusing.
Authors' response: Please note that the log y-scale is percent, not fraction. At a depolarization of 0.2 the percent of particles is ~1 %, not 100 %. Note that the log y-scale is percent, not fraction.
Authors' changes in the manuscript: None.

Referee Comment: Fig 6: Does it make sense to include the error bar information in the caption to make the figure less busy? Or at least remove and caption some of it?
Authors' response: We agree that the figure is busy, but the error bars reported are a big part of our discussion so it's important to retain these in the figure.
Authors' changes in the manuscript: No change has been made.

Referee Comment: Pg 17, line 1: typo of added "u". Also here you switch from >< notation to larger than and smaller than.
Authors' response: The text has been modified as suggested. "Larger than" and "smaller than" are now used throughout the text.

Referee Comment: Pg 18, line 12: typo likely

Authors' response: Corrected.

Referee Comment: Pg 20, line 4: double check wording for how this figure is introduced and also in the caption to be consistent and correct
Authors' response: The figure has been modified and now labels M as the "multiplication factor" as stated in the text and caption.

Referee Comment: Pg 20, line 5: suggest figure or Fig. 7
Authors' response: Corrected to "Figure"

Referee Comment: Fig 8: Caption could be improved, especially repetition in description of panel c.
Authors' response: Agreed and corrected.
Authors' changes in manuscript: The caption now reads "Figure 8: Application of depolarization ratio method on three CFDC runs. Aerosol composition and temperature are labeled in the title. (a) Time series of supersaturation with respect to water. (b) INP Concentrations under normal (blue) and WDBT (red) conditions are shown for the traditional (circles) and new (asterisks) analysis methods. (c) The normalized number distributions of all particles detected by the CASPOL. Time is reported in local time (CET)."

Referee Comment: Pg 20, line 26, suggest: In 2 out of 3 cases shown. Alternatively, you may want to clearly state (as you do later) that 27 cases/periods were evaluated (see Fig 9).
Authors' response: This is a good suggestion. Done.

Referee Comment: Pg 21, line 7: center panel of Fig. 8c?
Authors' response: Changed to "middle" panel.

Referee Comment: Pg 21, line 12: is data missing from fig 9 or is it just hard to see?
Authors' response: We have confirmed that no data is missing here. There are cases where no WDBT conditions occur in a run so there is no data to report.
Authors' changes in the manuscript: On page 22 ln 5, this is now noted in the manuscript, "In cases 24, 25, and 26 WDBT did not occur, so no data is reported.

Referee Comment: Fig 10: Which axis contains the data for the new method? I suspect the x, but am unsure due to confusion noted above. Please clarify and update axes. Would it make sense to fit this data to observe is there is a small bias in the new data?
Authors' response: The figure has now been correctly labeled with the traditional concentration on the x-axis and the new concentration on the y-axis. Though a fit could be used to describe the bias, the authors felt that a new discussion about a completely different application of a linear regression would be confusing to readers. The object of the plot is achieved by discussing the biases of the new method.

Referee Comment: Pg 22, paragraph beginning on line 5: I am confused about how the errors in two regions can be 500 and 50% but overall it's 32%. I believe this is averaged values for each region considered. Is this the best way to present the uncertainty? Also as a minor detail, spacing

when reporting numbers is inconsistent here and somewhat throughout the document, which would probably be fixed upon typesetting.
Authors' response: Fit to the results of the linear regression (Eq. 9), which has a very large y-intercept contribute to this variable performance. The 500% represents the lowest range of concentration detected and represents just a small portion of the large range of INP concentrations measured during the FIN-02 campaign. Above the 50,000 L^{-1}, the new method's performance improves greatly with measurements are within 50 % of the traditional concentration. No measurements here have error larger than 50%. The mean error for all measurements is 32.1 %.
Authors' changes in the manuscript: We've reordered and modified the text slightly to make this clearer.

Referee Comment: Figure 11: Consistency with previous figures and also double check captioning.
Authors' response: Thank you. The caption is now consistent with other figures.

Referee Comment: Pg 23, line 9: Does Fig 11b warrant more of a discussion? Can a literature comparison be included?
Authors' response: This case was discussed earlier in the text, and does deserve an additional brief discussion here. Since a detailed FIN-02 intercomparison is forthcoming in the literature in the near future (DeMott, et al, 2017), we limit the discussion here.
Authors' changes in the manuscript: This text (p. 26 ln 6) now reads, "Fig. 11b shows the special case of high activation of INP discussed above and in Figure 8b. This case involves a highly active INP, Snomax® at -20 ˚C, a significantly colder temperature than required for the Snomax® to activate as INP. Since most particles activated prior to the onset of WDBT, there is negligible difference in the concentrations reported during "ice only" and WDBT periods."