Authors' response to Anonymous Referee #1 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 #1 Received and published: 9 August 2017
Referee Comment:
Review of "Using depolarization to quantify ice nucleating particle concentrations: a new method" by Zenker et al.
General Comment This manuscript introduces a new method to distinguish between ice particles, aerosol particles, and liquid water droplets at the water droplet breakthrough (WDBT) line in a continuous flow diffusion chamber. The traditional method to determine the concentration of ice nucleating particles (i.e., particle size) is not accurate at the WDBT and therefore, the proposed method can be of high importance. The new proposed method agrees well with the traditional method before the WDBT and it improves the detection of INPs at and above this line. However, this new method cannot be applied to field measurements given that the uncertainty is very high when low concentrations of INPs are present. Therefore, this new method is only valid for laboratory experiments where high concentrations of INPs are usually achieve. Although the scientific goals are interesting and the experiments/analysis were carefully performed, the presentation of the manuscript is not the best. There are too many typos, some parts are repeated along the manuscript, and there are key references missing. It would be nice if a senior researcher from the team can proof-read the revised version. **The reviewer did not find a major point; however, the following minor comments need to be addressed before its publication in AMT.**

Authors' response: Thank you. As the reviewer notes, we have stated in the original text that this technique is best applied to laboratory measurements due to signal to noise. Nevertheless, we agree with the reviewer that developing such a method is of high importance due to the need for such a method, and we thank the reviewer for supporting this work.
Authors' changes in manuscript: The manuscript has been carefully revised and the minor comments of the reviewer are addressed item by item below.

Minor Comments
Referee Comment:
P2 L6: "depositional freezing" is incorrect given that "freezing" refers to the transition from liquid to solid. In deposition ice nucleation the liquid phase is not present.
Authors' response: Corrected.

Referee Comment: P2 L9-10: In all clouds or Mixed-phase clouds only?

Authors' response: In general, immersion freezing is the dominant nucleation mechanism for producing ice crystals in all clouds containing ice.

Referee Comment: P2 L17: Why mixed-phase clouds exclusively? Heterogeneous ice nucleation can also takes place in cirrus clouds, for example.
Authors' response: We did not mean to imply that ice nucleation mechanisms are only relevant to mixed phase clouds. Mixed phase clouds are mentioned here because the current study addresses the specific challenges of accurate detection of ice in the presence of droplets.

Referee Comment: P2 L18: Add references after "GCMs". P2 L20: There are many studies showing this. I will rather cite a review paper instead.
Authors' response: Added "(e.g. Tan et al., 2016; Pithan et al., 2014)".

Referee Comment: P2 L22: Atkinson et al. (2013) and Yakobi-Hancock et al. (2013) are not field studies.
Authors' response: True. We have changed "Field measurements" to "Measurements".

Referee Comment: P2 L24-25: Other groups working on ice nucleation (besides the two cited here) have done a significant contribution as well. It would be better to divide the references by aerosol type. The recent reviews by Coluzza et al. (2017) and Kanji et al. (2017) nicely fit here.
Authors' response: This is certainly true. We were focusing on the TAMU CFDC which is the topic of the manuscript and the CSU CFDC to which is it compared in this manuscript. But, we agree that other importance contributions should be mentioned.
Authors' changes in manuscript. We now include Coluzza et al. (2017) and Kanji et al. (2017).

Referee Comment: P3 L8: Add references after "crystals".
Authors' response: Reference to Bohren and Huffman, 1983 added.

Referee Comment: P3 L16: The Cziczo et al. (2017) review could be cited here.
Authors' response: Done.

Referee Comment: P6 L13: What do the authors mean with "processing chamber"?
Authors' response: Please refer to pg. 6 ln 4 which now reads "The aerosols then enter the CFDC processing chamber where temperature and supersaturation are controlled."

Referee Comment: P6 L14: Remove "TAMU". It was previously mentioned that CFDC will refer to the TAMU CFDC.
Authors' Comment: Done.

Referee Comment: P7 L4: Why were the flows changed? Should it not be constant?
Authors' response: Flow adjustments were made to maintain conditions within the critical flow regime, i.e. to ensue laminar flow, and avoid buoyancy effects under all operating temperature and supersaturations. Ideally, these flows would be constant; however in order to obtain the certain high supersaturation targets which were of interest during FIN02 evaluation of WDBT, adjustments were made.

Referee Comment: P7 L4: Please add the uncertainty for 2 L min-1.

Authors' response: Note that experimental uncertainty is not discussed in this sentence. This is variation in selected operating flows (±0.5 L min^{-1}) during FIN02. In contrast, uncertainty in sample flow is ±0.1 L min^{-1} based on the experimental precision in the total and sheath mass flow controlled.
Authors' changes in manuscript: Added "±0.5 L min^{-1}."

Referee Comment: P7 L4: I found the 1.5°C value quite high. Other CFDC report much lower values. What is the reason for this?
Authors' response: This is not an instrumental uncertainty, per se. It is a choice of data processing. All CFDC data collected at a mean temperature of X ±1.5°C was included in the data set for a chosen temperature X.

Referee Comment: P7 L10: Remove "and" after "pressure".
Authors' response: Done.

Referee Comment: P7 L15-16: "The concentration of particles measured while the filter is in place is subtracted from the total concentration measured by the CASPOL." Both are measured by the CASPOL. I think it would be better to say: Total concentration measured during the supersaturation scan. To account for this, a filter is placed ahead of the sample inlet in order to determine background signal of the CFDC chamber. The background period that is closest to a given 1-minute sample period is then applied by subtracting that background concentration from the total concentration measured by the CASPOL at the sample time.
Authors' response: We don't see exactly how the reviewer's suggestion would work, but we have revised the sentence for clarity.
Authors' changes in the text: The text now reads, "… a filter is placed ahead of the sample inlet in order to determine background signal of the CFDC chamber. The background period that is closest to a given 1-minute sample period is then applied by subtracting that background concentration from the total concentration measured by the CASPOL at the sample time."

Referee Comment: P7 L18: Add references after "crystals".
Authors' response: Added Bohren and Huffman, 1983.

Referee Comment: P7 L28: "Any droplets that remain larger than the 2 μm size cut will be miscounted as ice". This is based on who?
Authors' response: We are not certain what the Referee means to ask here. We feel it is clear as stated that when the size-cut is set to 2 μm, any particles, be they ice or other composition, which are larger 2 μm will be counted as ice.

Referee Comment: P8 L7: "discern". Between what?
Authors' response: Changed to "determine".

Referee Comment: P8 L8: What do the authors mean with positive and negative artifacts?
Authors' response: This was a mistake on our part, because only positive artifacts are possible. "Positive artifacts" mean water droplets breaking through are counted as members of the ice

crystal population. "Negative artifacts" would mean ice particles not counted because one thinks they are water droplets, but in practice there is no way for that to occur.
Authors' changes in manuscript: "if the instrument is unintentionally operated at supersaturations above WDBT, droplets will be miscounted as ice crystals."

Referee Comment: P8 L11: This sound a bit awkward.
Authors' response: The original sentence was: "For the traditional analysis method to be successful, sample aerosols must not be larger than the applied size cut or they too will be miscounted as an INP."
Authors' changes in manuscript: Revised to: "In the traditional analysis, any sample aerosols larger than the applied size cut will also be miscounted as INPs."

Referee Comment: P8 L15: Add references after "signal".
Authors' response: This is based on empirical testing. There is no literature reference necessary.

Referee Comment: P8 L15: This is the fourth time the word "new" is used.
Authors' response: The topic of this manuscript is to compare one method, the "new method" developed here to the "traditional method", so we need to reserve the right to say "new" many times.

Referee Comment: P8 L16: What is "high" and "low"?
Authors' response: In the revised version of the document this sentence has been deleted due to other suggested revisions.

Referee Comment: P8 L16: Replace "our" with "the".
Authors' response: Removed "our" in the text.

Referee Comment: P9 L4: Add reference after "infinite"
Authors' response: This is a straightforward mathematical interpretation of Equation 2 above it. We do not feel it needs a reference.

Referee Comment: P9 L11: Delete "Using" after "…droplets."
Authors' response: Done.

Referee Comment: P9 L15: Add references for the 1.33 value.
Authors' response: Added Zajak and Hecht, 2002.

Referee Comment: P9 L16: Add the uncertainty for the droplet sizes.
Authors' response: Added.
Author's change in the manuscript: "As reported in Glen and Brooks (2013), the uncertainty in sizing due to differences in the complex refractive indices of oil and water are up to 30% based on a comparison of VOAG oil droplet calibrations of CASPOL to water-based calibrations performed by the manufacturer. For this project, droplets were generated with the diameters of 2 ± 0.6 μm, 6 ± 1.8 μm, 8 ± 2.4 μm, and 10 ± 1.5 μm."

Referee Comment: P9 L18: Remove "and" after "frequency".

Authors' response: Here the grammar is correct as written.

Referee Comment: P9 L22: “sample flow is split between flow to the CASPOL” sound a bit awkward. Remove one “the”.
Authors' response: Done.

Referee Comment: P9 L22: Remove “and” after “controller”.
Authors' response: Here the grammar is correct as written.

Referee Comment: P9 L24: Replacer “are” with “were”.
Authors' response: Done.

Referee Comment: P10 L4: “in aerosols”?
Authors' response: Changed to "of aerosols"

Referee Comment: P10 L12-23: “in the absence of activated liquid droplets”. Do the authors mean in the absence of INPs?
Authors' response: No. The referee is correct that homogenous freezing occurs without INP. However, the point we're making here is that the experiment under cold, dry temperatures to reduce the chance of any unwanted droplet formation.

Referee Comment: P10 L15: “-11 ± 1.5 % SSw”? Something is wrong here.
Authors' response: We have rearranged and simplified the sentence for clarify.
Authors' changes in manuscript: The text now reads, "the CFDC was operated at –55 ± 0.2 °C and 51 ± 2.3 % SS_i(-11 ± 1.5 % SS_w)..."

Referee Comment: P10 L26: Remove “TAMU”. See comment on P6
Authors' response: Done.

Referee Comment: L14 P11 L16: “Fig.s 1” should be “Fig. 1”.
Authors' response: Fixed.

Referee Comment: P12 L17 and P13 L6-8: Why did the authors choose dust-like as the model for aerosol particles? How about biological particles? Soot?
Authors' response: Indeed, aerosols come in a wide variety of compositions. Performing scattering calculations on all atmospheric aerosol types would be beyond the scope of this study. We chose dust as a relevant choice due to its widespread presence in the atmosphere and the known action of many dusts as INPs. Also, many of the organic and inorganic salt particles in the atmosphere will be in the form of solution droplets. Since those will have similar scattering properties to the water droplets, which were already included, the non-spherical dust also provides a good compliment to the spherical particles.

Referee Comment: P13 L27: Please indicate to what Figure the authors are referring to.
Authors' response: Fig. 1a is now stated.

Referee Comment: P14 L23-24 and along the manuscript: Please use "WDBT" instead of "water droplet breakthrough". This was defined in P8 L1.
Authors' response: True, but we find it helpful to write it out in full one more time for readers who are likely to be unfamiliar with the term.

Referee Comment: P15 L5: Please indicate to what Figure the authors are referring to.
Authors' response: Fig 1a is now included.

Referee Comment: P16 L24-26: Replace "um" with "μm" to be consistent.
Authors' response: Corrected.

Referee Comment: P17 L8: I think the year of the Pruppacher and Klett book is incorrect.
Authors' response: Thank you, we have corrected this oversight.

Referee Comment: P17 L23-24: "the geometry of the ice crystal can be modified leading to drastic differences in the observed depolarization ratio." Can the authors report the time scale under which this is valid? i.e., how many seconds/minutes are needed for an ice crystal to change its geometry?
Authors' response: This comment pertains to the study of Smith, 2016. . They grew ice crystals in the Manchester Ice Chamber which is similar in size and design to the AIDA chamber. They operated the chamber at various temperatures and grew ice crystals in the chamber. The change in relative humidity reportedly causes the evolution of ice crystal geometry. To monitor the changes, sample were taken every minute over 5-6 minutes. There were visually noticeable changes in to the habit at each minute increment (e.g. hollow hexagonal columns evolve slowly into solid hexagonal columns, dendrites evolve into hexagonal plates). The paper does not report a specific timescale that is necessary for a detectable change in ice crystal shape to occur, but their results suggest that changes may occur rather rapidly with changing conditions in the chamber (over a minute or less). While this is an interesting study, we feel it is beyond the scope of our study and interested readers are referred to the Smith paper.
Authors' changes in the manuscript: None.

JAKE Referee Comment: P17 L24 and 27: Add the year of the Smith et al. paper.
Authors' response: The year is now in the text "Smith et al. (2016)"

Referee Comment: P17 L28: "(2016)" is out of place.
Authors' response: Corrected.

Referee Comment: P18 L2: Add "field" before "campaign".
Authors' response: FIN-02 was a laboratory campaign, FIN-03 was a field campaign. These are now correct everywhere in the text.

Referee Comment: P19 L25: Please indicate to what Figure the authors are referring to.
Authors' response: Fig. 7 is now mentioned in the text.

Referee Comment: P22 L3-4: How about to include kanji et al. (2017)?
Authors' response: Good idea. Done.

Referee Comment: P22 L8: "the Colorado State University (CSU) CFDC". This was defined already in P3 L18.
Authors' response: True, but in discussion with coauthors we decided a little repetition for clarify was helpful.

Referee Comment: P23 L19: Add "only" after "experiments".
Authors' response: Done.

Referee Comment: P25-30: Be consistent with the journal names in the references. Either add the full name or their abbreviation.
Authors' response: Abbreviations are now used in accordance with the AMT manuscript preparation guidance.

Jake Referee Comment: P25-30: The page numbers in several references are missing (e.g., DeMott et al. (2017), Levin et al. (2016), McCluskey et al. (2016), McFarquhar et al. (2011)).
Authors' response: All page numbers are now included where applicable.

Referee Comment: P25-30: References need to be up to date.
Authors' response: We have added a few references from 2017 per the reviewer recommendations (Coluzza et al., 2017; Cziczo et al., 2017; Kanji et al., 2017).

Figure 2: Given that there is no extra-charge for colored-figures in AMT, I suggest to add color to this figure to improve its readability.
Authors' response: Since the symbols and colors on Fig. 2 were chosen to match the same data sets included on page. 6, we prefer to keep the original scheme.

Referee Comment: Figure 3: Blue circles in panel's b and c should be blue squares.
Authors' response: This has now been fixed.

Referee Comment: Figure 11: "TAMU CFDC versus CSU CFDC comparison."
Is written twice in the figure caption.
Authors' response: Corrected. Thank you.

Referee Comment: Table 1: Add ":" after "1" for consistency with the Figures.
Authors' response: Done.

References:
Coluzza, I., Creamean, J., Rossi, M. J., Wex, H., Alpert, P. A., Bianco, V., Y. Boose, C. Dellago, L. Felgitsch, J. Fröhlich-Nowoisky, H. Herrmann, S. Jungblut, Z.A. Kanji, G. Menzl, B. Moffett, C. Moritz, A. Mutzel, U. Pöschl, M. Schauperl, J. Scheel, E. Stopelli, F. Stratmann, H. Grothe, and D. Schmale III (2017). Perspectives on the Future of Ice Nucleation Research: Research Needs and Unanswered Questions Identified from Two International Workshops. Atmosphere, 8(8), 138.

Cziczo, D. J., Ladino, L., Boose, Y., Kanji, Z. A., Kupiszewski, P., Lance, S., Mertes, S., and Wex, H. (2017). Measurements of Ice Nucleating Particles and Ice Residuals. Meteorological Monographs, 58, 8.1-8.13.

Kanji, Z.A., Ladino, L., Wex, H., Boose, Y., Burkert-Kohn, M., Cziczo, D.J., and Krämer, M. (2017). Overview of Ice Nucleating Particles. Meteorological Monographs, 58, 1.1-1.33