Authors' response to Anonymous Referee #3 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 #3 Received and published: 9 August 2017

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. 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.

Authors' response: Thank you. We agree that WDBT needs to be addressed for ice nucleation measurements to be more accurate and performed reliably under a broader range of atmospherically relevant conditions. Also we agree that depolarization ratio can improve our discrimination between INP and wayward droplets reaching the detector, as our manuscript shows for the laboratory experiments herein.
Authors' changes in the text: We have restructured the introduction, moved the section on particle depolarization, and significantly revised the data analysis and interpretation sections, as well as the modeling section, as discussed in the point by point response and in response below

and in response to the other reviewers. We feel that the manuscript has improved greatly in readability and clarity and hope that the referee agrees.

General Comments:

Referee Comment:
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.
Authors' response: We have restructured the introduction, following the later comment of the reviewer that the benefit of the new methods does become clear on page 4. We have moved that text forward to earlier in the introduction. We do note that current phase discrimination studies available in the literature are cited, and there are not a very large number available, which is further reason that we'd like to see the present study published.
Authors' changes in the manuscript: Please see the revised introduction.

Referee Comment: 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.
Authors' response: Section 2 has been restructured, with subsections 2.1 and 2.2 reordered (switched), and has been revised according to specific Referee comments here and in the other reviewers. As cited in Section 2, the TAMU CFDC and CASPOL have been discussed in great detail in our previous work (Glen & Brooks, 2013 and 2014, and Glen, 2014.)
Authors' changes in the manuscript: Please see the revised section 2 in the text.

Referee Comment:
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.
Authors' response: In section 3.3, the training data sets are introduced. In Figure 3B, the population of particles is plotted against depolarization ration. It can be seen from this figure that droplets have depolarization ratios up to 0.3. Therefore, we visually assign 0.3 as the nominal depolarization threshold cut-off. At this point, the caveat remains that a small percentage of aerosols do have depolarizations greater than this threshold. However, in the case of aerosols, there are 2 lines of defense against any aerosols being accidently counted as ice nucleation. In addition to the majority of aerosols having in addition to the depolarization threshold, aerosols with sizes above 1.75 micron diameter are physically removed from the sample upstream of the CFDC chamber.

In section 3.7, the linear regression model is introduced and used to optimize the cut-off. The statistically significant results in confirm that 0.3 is an optimal choice of threshold.
Authors' changes in the text: For clarity, section 3.3 now includes the statement. "For each training data set, the frequency distribution of depolarization ratio reported as a percentage of the

total particles in the data set is shown in Fig. 3b. It can be seen from this figure that droplets have depolarization ratios up to 0.3. Therefore, we visually assign 0.3 as the nominal depolarization threshold cut-off for differentiating between ice crystals and non-ice particles. Unfortunately, it can also be seen that a small percentage of aerosols do have depolarizations greater than this threshold.   However, since aerosols with sizes above 1.75 micron diameter are physically removed from the sample upstream of the CFDC chamber, the combined consideration of size and depolarization may prove a robust strategy for avoiding the miscounting of aerosols as INP as further discussed below."

Secondly, based on referee comments, section 3.7 has largely been rewritten.

Referee Comment: 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.
Authors' response: Linear regressions are commonly used when determining how to use a new technique to determine an atmospheric quantity by relating a measured parameter or parameters to a ground truth measurement.
Authors' changes in the text: To explain to the user that there is a wide array of applications for a linear regression, several more papers that use a linear regression for various purposed (Zimmerman et al. 2017; Brunner et al., 2015; Choi et al., 2016) are now cited.

Referee Comment: Also expand on how the choice of another depolarization ratio threshold does influence your results.
Author response: Please see the rewritten section 3.7

Referee Comment: 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.
Authors' response:
The TAMU and CSU CFDCs are independent custom-built instruments which operate on the same principles. Most importantly, because the CSU CFDC doesn't experience WDBT until a higher RH, this comparison provides a means to evaluate performance of the new method under conditions which our traditional method is clearly failing. In general, because the two CFDCs differ in dimensions, flow rates, operating conditions (temperature and supersaturation) in the growth and evaporation regions within the instrument, and the choice of detector and size cut-off, an intercomparison is worthwhile.

Most importantly, because the CSU CFDC doesn't experience WDBT until a higher RH, this comparison provides a means to evaluate performance of the new method under conditions which our traditional method is clearly failing. Also, in general, because the two CFDCs are quite different instruments, an intercomparison is worthwhile.

We emphasize that the choice of different size cuts are justifiable, because the two CFDCs are not the same instrument in some key regards.   For example, the TAMU CFDC growth chamber is smaller and the residence time is shorter. Therefore ice particles are not expected to grow as large as in the CSU unit. Logically, a small size cut may be more appropriate for the TAMU instrument. However, there are competing parameters, making this a non-straightforward choice, which is why multiple size cuts have been included for consideration in Fig 11. This is an issue that any operator of this type of instrument is concerned with.

Authors' changes in the manuscript. For emphasis we include the following statement on pg 24, ln 20, "Thus, inclusion of the CSU data provides a test of the new method at higher relative humidities under conditions when data obtained through the TAMU CFDC's traditional method is clearly spurious due to water droplet breakthrough."

Also, regarding differences in the 2 CFDCs, on pg 9, ln 14-20, the text now reads, "CFDCs in use today are custom-built instruments which vary in physical dimensions and choice of detector, although all operate under the same basic principles. Due to the combination of different chamber dimensions, flow rates, operating conditions (temperature and supersaturation) in the growth and evaporation regions within the instrument, and the choice of detector and size cut-off, WDBT varies from instrument to instrument."

Referee's comment: 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.

Authors' response:
The specific conditions of WDBT vary with CFDC temperature, the ambient humidity, the hygroscopicity of sample aerosols, the size of sample aerosols, and the sample flow which determines the residence time in the instrument.
Author's changes in text: This is now stated in the text on page 9, ln 9 as written, "Specific conditions of WDBT vary with CFDC temperature, the ambient humidity, and the hygroscopicity of sample aerosols, the size of sample aerosols, and the sample flow which determines the residence time in the instrument. Typically, in the TAMU CFDC the onset of WDBT occurs at 3 % to 4% SS_w, but has been observed as low as 1 % SS_w and as high as 8 % SS_w."

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). However, the succinct discussion of ice nucleation mechanism and mixed phase clouds are integral parts of the discussion on the topic of INP so they are not been removed.

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. 1625). 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.

Authors' response:
The introduction has been significantly revised with the section on particle discrimination moved to early in the introduction.

However, given that this is a study on improvements in ice nucleation instrumentation, we feel the historic details lend important context to the issues, especially those related to water droplet breakthrough and the improvements our new methods contributes.
Hence, we keep the majority of the text and we have added additional details specifically on strategies instruments through history have used to differentiate between ice crystals, water droplets, and aerosols. Also, we would be remiss to leave out (or delete) the section defining ice nucleation mechanisms, so that section remains.

Traditionally phase discrimination has relied on differences in particle size. An impactor is used to physically eliminate aerosols larger than a certain point (~ 1.75 micrometer diameter). Traditional detectors are optical particle counters which detector particles in a range of sizes.
Author's changes in text: Please see the revised and reorganized introduction.

Referee Comment:
P. 4 l. 413 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 reference s here as well.
Authors' response: Please see our response to the next comment below. It appears that as the reviewer read further s/he found the answers to his/her questions. To make things clear sooner, we have reordered Sections 1 and 2 of the text, as discussed above.

Referee Comment:
Finally, the benefit of new methods, as described in the presented study becomes clearer.
Authors' response: Thank you. We are glad that this section clarified our motivation for new method development, and have moved that section forward in the text.

Referee Comment:
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.
Authors' response: Please see above in response to specific changes we have made.

Referee Comment: P. 1, l. 12: Please change “observed” to “measured”.
Authors' response: The text is unchanged.

Referee Comment: 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."
Authors' response: Okay.
Authors' changes in the manuscript: The text now reads," During WDBT, the standard procedure of counted counting all particle which grow beyond the size cut-off as ice crystals fails, which large droplets are miscounted as ice."

Referee Comment: 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.
Authors' response: Okay.
Authors' changes in the manuscript: Revised to read, "To accurately measure INP during WDBT..."

Referee Comment: P. 1, l. 25: Please change "complicated" to "complex".
Authors' response: We prefer the original. The text is unchanged.

Referee Comment: P. 1, l. 26: Please clarify whether "precipitation" refers to spatial/temporal distribution of precipitation, precipitation formation or precipitation in general
Authors' response: As written, precipitation in general is implied,.. "Because of their complicated microphysical properties, ice clouds and mixed-phase clouds pose challenges in understanding our global radiative budget and precipitation."

Referee Comment: P. 2, l. 2: Leave out "our".
Authors' response: Done.

Referee Comment: P. 2, l. 8: Leave out "becomes"
Authors' response: Done.

Referee Comment: P. 2, l. 11: Please insert: "aerosol particle…"
Authors' response: Done.

Referee Comment: P. 2, l. 12: Please change to: "aerosol particle collides with a supercooled water droplet and …"
Authors' response: Since, as the next sentence in the text states, "While the exact mechanism of contact freezing remains unresolved, it has been shown that the presence of an INP positioned at a droplet surface facilitates freezing at temperatures several degrees warmer than immersion freezing with identical INPs (Fornea et al., 2009; Durant and Shaw, 2005).", we feel the original is more accurate.

Referee Comment: P. 2, l. 20 : Delete "Field"
Authors' response: Done.

Referee Comment: P. 2 , l.23: Delete “other”
Authors' response: Done.

Referee Comment: P. 2, l. 28: Delete “can”
Authors' response: Done.

Referee Comment: P. 3, l. 4: Please change to: “…components of the LIDAR signal retrieved from…”

Referee Comment: P. 3, l. 1315: 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.
Authors' response: We thank the Referee for pointing out that the first sentence here was our place in this paragraph. It has been moved to the optical section of the introduction.

Referee Comment:
P. 3, l. 1022: Shorten this paragraph and to keep the focus on the topic of your manuscript.
Authors' response: As stated above, the introduction is significantly revised and rearranged. This paragraph no longer exists in its original form.

Referee Comment: P. 3, l. 23: Please change to “… (CLIMET Inc., Model No. CI3100) …”
Authors' response: Done.

Referee Comment: P. 4, l. 13: Delete “to detect INP”
Authors' response: Revised to, "to determine INP concentration."

Referee Comment:
P. 5, l. 4: Please change to: “… are generated, suspended in dry synthetic air…”
Authors' response: Done.

Referee Comment:
P. 5, l. 7: Please specify whether aerosol particles or populations of ice crystals and cloud droplets have been sampled from AIDA.
Authors' response: Thank you. This is an important distinction.
Authors' changes in the manuscript: Revised to read, "During FIN-02, prior to expansion, aerosols were drawn from the AIDA chamber by the various ice nucleation instruments. Following the aerosol sampling period, an AIDA expansion was performed so that INP concentration determined by AIDA could be compared to results from the various visiting instruments."

Referee Comment: P. 5, l. 9: Please add: “… of the TAMU CFDC-CASPOL measurements …”

Authors' response: Done.

Referee Comment: P. 5, l. 25: Please change “limited” to “small”.
Authors' response: Done.

Referee Comment:
P. 6, l. 14: 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?
Authors' response: As cited in the text, experimental details and a full description of the development and characterization of the TAMU CFDC are provided in the references below:
Glen, A., and Brooks, S.D.: Single particle measurements of the optical properties of small ice crystals and heterogeneous ice nuclei, Aerosol Science and Technology, 48(11), 1123-1132, 2014.
and
Glen, A.: The development of measurement techniques to identify and characterize dusts and ice nuclei in the atmosphere. Diss. Texas A&M University, 2014.

Referee Comment:
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.
Authors' response:
By definition, when droplets only partially evaporate, the chamber is under WDBT conditions. Causes on WDBT have been discussed in detail above and earlier in the text. Ideally, no droplets should survive the evaporation region of the instrument, but given that WDBT is a problem in this and many other ice chambers, we see that in practice this is not the case. There are many possible reasons. For instance, droplets may not come to equilibrium prior to existing the chamber under very moist ambient conditions.

CFDC flow conditions were already stated in the original text on page 7. "Two mass flow controllers are used to set the total flow and recirculating sheath flow through the chamber. The difference between the total and sheath flows determines the sample flow. For this campaign, the total flow was set to values ranging from 6 to 9 L min^{-1} and the sheath flow was set to values ranging from 4 to 7 L min^{-1} resulting in a sample flow that was typically ~2 ± 0.5 L min^{-1}."

In Figure 6, direct CFDC measurements are reported. The particles detected (not implied) by the CFDC do include 40 micron diameter particles in size. Ice growth calculations indicate that ice crystals may grow rapidly in size in the chamber (Rogers, 1988; Glen, 2014). Additionally, a known source of large ice crystals are shards that break off the chamber walls occasionally.
Author changes in manuscript: Changes referred to here are all parts of the revision discussed in reference to prior comments.

Referee Comment: 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.
Authors' response: We respectfully disagree. When a manuscript employs an instrument and experimental procedure which are previously published in detail, there is always a delicate balance between re-reported what has been well documents in previous work or not providing enough basic details for a reader to follow the current manuscript. In this case, the referee has asked for additions experimental details above and here asks for fewer details. We do not think it wise to remove the details included in the original.

Referee Comment: P. 6, l. 17: Please change to: "… (CLIMET Inc., Model No. CI-3100)…"
Authors' response: Done.

Referee Comment: P. 6, l. 26: Please change to: "…backward scatter detector…"
Authors' response: Done. Thanks for pointing this out.

Referee Comment: P. 6, l. 5: The position of the mass flow controllers should be specified. I assume these are located downstream of CASPOL?
Authors' response: For clarity, the text has been revised. For a schematic, please see Glen and Brooks, 2014a.
Authors' changes in the text: The text on page 7 ln 14016 now reads "...the CASPOL is installed at the base of the chamber. Two mass flow controllers downstream of CASPOL are used to set the total flow and recirculating sheath flow through the CFDC-CASPOL. The difference between the total and sheath flows determines the sample flow."

Referee Comment: P. 7, l. 13: Please change to: "Temperature, …"
Authors' response: Done.

Referee Comment: P. 7, l. 17: Please change "ahead" to "upstream"
Authors' response: Done.

Referee Comment: P. 7, l. 119: 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.

Authors' response: Okay, the text is now clarified as below. In our experience, RH doesn't appear to cause a large difference in background signal.
Author changes in manuscript: The text now reads "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: 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 cutoff 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” refer s to. *Authors' response:* Yes, thanks. We see how this could have been confusing in the original text. Actually, we refer to any size-discrimination (by OPC or CASPOL forward scatter detector) as traditional analysis, and the use of the depolarization as a new method.

Author changes in manuscript:

p. 4 ln 12: "Particles are sized according to the intensity of light which reaches the CASPOL's forward scatter detector, as in a traditional OPC."

and

p. 8 ln 6: "During FIN-02, data collected by the CASPOL's forward scattering detector was used for the traditional analysis."

Referee Comment:

P. 7, l. 27: This statement is misleading. There are no limitations of the OPC technique (dis crimination 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.

Authors' response: For a detailed discussion of the many causes of WDBT and related instrument details, please see page 8 ln 14-22, which have been expanded and revised. Note, however, that we do not refer to thermodynamic conditions, however, because WDBT is consistent with failure to remove large supercooled drops which do not reach thermodynamic conditions by the time they reach the chamber.

Referee Comment:

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.

Authors' response: Ice crystal size thresholds have been chosen empirically based on laboratory results. For further details please see our previous work (Glen, 2014B).

Referee Comment: P. 8, l. 8: What do the authors mean by positive or negative artifacts?

Authors' response: This was a mistaken choice of words, as noted by 2 referees. In reality 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:
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.
Authors' response: This is the second time the referee mentions this issue, which is really a word choice issue. We revised the language in the abstract as per his/her recommendation. For clarity, we feel it best to keep the original text here, as this manuscript is addressing a measurement challenge specifically occurring when WDBT occurs.

Referee Comment: 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.
Authors' response: Given the importance of this issue, we keep this section on page 8, but have shortened it.
Authors' changes in manuscript: Pg 8 now reads, "Although operation with an upstream impactor reduced this problem, ~1 to 10% of particles larger than 2 μm (depending on flow) may make it into the chamber to contribute to the apparent INP signal."

Referee Comment: 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?
Authors' response: At this stage in the manuscript, the text only states that such capabilities would be an improvement, given the limitations of the traditional analysis.
Authors' changes in manuscript: The text now reads, "A new analysis method that differentiates between large aerosols and ice crystals is needed since it would remove the need to limit the size of particles allowed into the instrument in the first place."

Referee Comment:
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 t he statement in brackets on p. 8, l. 10.
Authors' response: The statement in question was deleted in response to the Referee's previous comment.

Referee Comment: I recommend moving the discussion of section 2.4 to the Introduction to motivate the development of the new method.
Authors' response: Please see above that we have significantly revised the introduction. Moving this particular section was something the authors previously discussed. We decided that so much detail about challenges specific to our CFDC would be better left in the experimental section.

Referee Comment:
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.
Authors' response: We see the referee's parts 1 and 2 as parts of the same objective. We prefer to keep the original text here.

Referee Comment:
P. 9, l. 1: I suggest repeating the meaning of the different parameters again. E.g. “Similar to eq. (1) B□,CAS and B||,CAS denote the perpendicular and parallel components of the backscattering signal, respectively, and the subscript CAS refers to the CASPOL signal….”
Authors' response: This is a good suggestion.
Authors' changes in manuscript: Done.

Referee Comment:
P. 8, l. 24: Section 2.5 describes CASPOL instrumental details and should be moved to the description of CASPOL in section 2.2.
Authors' response: This is a good suggestion. Due to other suggestions the CASPOL description is now in Section 2.2. and Section 2.5 has been moved to that section.

Referee Comment: P. 9, l. 21: Please explain why the neutralizer prevents particle loss.
Authors' response: Charged particles are attracted to the walls of the tubing.
Authors' changes in manuscript: This statement is now including in the text..."to prevent particle loss since charged particles tend to be attracted to the walls of sample tubing."

Referee Comment: P. 9, l. 22: Please change to: “the” before CASPOL
Authors' response: No. The grammar is correct in the original.

Referee Comment: P. 10, l. 15: I assume you are referring to CFDC-CASPOL measurements,
Authors' response: Yes.
Authors' changes in manuscript: Changed CASPOL to CFDC-CASPOL.

Referee Comment: 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?

Authors' response: This is reported instrument uncertainty, whereas on 7, we reported the range of temperatures over which collected experimental data was included in the intercomparisons. Specifically, temperature here is based on experimental temperature, derived from a set of 8 thermocouples calibrated to a reference RTD.
Authors' changes in manuscript: The text on pg 7 has now been clarified to explain that the temperature range on pg 7 was not a report of instrument uncertainty. Instead, it was the range of operating temperatures of measurements included in the FIN-02 intercomparison.

Referee Comment:
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.

Authors' response: We feel that supersaturation is useful because 0 % demarcates when water droplets may begin to form in the chamber. Also, SS is often used in ice nucleation papers.
Authors' changes in manuscript: None.

Referee Comment:
P. 10, l. 20: Do you suggest that particles smaller than 2 μm are not necessarily frozen?
Authors' response: 2 μm is the nominal size cut for ice. Both calculations and experimental tests have shown that if size nucleation in our chamber, ice will grow to above 2 μm (Glen, 2014.)
Referee Comment: P. 10, l. 23: Insert comma after "datasets"
Authors' response: Done.

Referee Comment: P. 11, l. 4- 5: Please clarify the usage of optical signatures by Hu et al. (2009) and how this relates to your study.
Author's response: Revised for clarity. Hu et al is a successfully example of using backscatter and depolarization data to determine cloud particle phase.
Authors' changes in the manuscript: "In an analogous method, optical signatures produced from CALIPSO satellite backscatter and depolarization data have been used to identify cloud phase (Hu et al.; 2009)."

Referee Comment: P. 11, l. 6: Delete "training"
Author's response: Training has a very specific meaning here, so we choose to keep it.

Referee Comment: P. 11, l. 13: Please clarify whether Dp refers to the optical diameter measured with CASPOL or to another diameter measured with another device.
Author's response: For clarity Dp has been replaced with diameter and revised the text as below:
Authors' changes in manuscript: "As discussed, the ice crystal and droplet training data shown in Fig. 1 only includes particles with optical diameters ≥ 2 μm and ≥ 1 μm, respectively.

Referee Comment:
P. 11, l. 17: Please extend your interpretation of why almost only ice crystals show high values f or B□/F and what that implies.
Author's response: Here we report a direct observation from data in the figure.
Authors' changes in manuscript: No change has been made.

Referee Comment: P. 11, l. 23: Insert point after "et al."
Author's response: Done.

Referee Comment:
P. 11, l. 24 : Please clarify why this is an empirical tool and how this affects the application to your data.
Author's response: The optical signatures are used to detect patterns in backscattering vs. depolarization plots for different particle types. By definition, these observed differences (if found) are empirical rather than theoretical.
Authors' changes in manuscript: No change has been made.

Referee Comment:
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?
Author's response: The Referee has a good point. This is a reality, not an assumption.
Authors' changes in manuscript: Deleted, "It is assumed that."

Referee Comment: 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.
Author's response: As clearly stated in the text- z is the direction on propagation of the incident CASPOL laser beam. See page 12, ln 24, "The CASPOL emits an incident beam that propagates along the z direction in the form." Also, schematics of the CASPOL have been previously published in Glen and Brooks 2013 & 2014.)
Authors' changes in manuscript: No change has been made.

Referee Comment:
It is not the CASPOL, but the laser diode of the CASPOL that emits the light.
Author's response: True, "laser" now added.

Referee Comment:
P. 12, l. 8: Please change to: "…line linking particle (position) and detection point."
Author's response: We feel that this would not be an improvement.

Referee Comment: P. 12, l. 12: Please insert commas: "… ratio, δModel, can …"
Author's response: Done.

Referee Comment: P. 12, l. 15-16: Please insert commas: "…matrix, Pij, the amplitude matrix, Sij, and the scattering cross section, Csca, …"
Author's response: Done.

Referee Comment: P. 13, l. 13: Please replace "vs." by "as a function of"
Author's response: Done.

Referee Comment: P. 13, l. 17: Please delete "the" in front of optical signatures.
Author's response: There is no "the" in the line specified.

Referee Comment: P. 13, l. 13-21: Please elaborate this discussion and give more details:

Author's response: This section has been rewritten and expanded. Also, the range of particle and ice diameters have been expanded in Figure 2,as discussed in the new text.
Authors' changes in manuscript. Please see the revised section on Pg 14 ln 15 to pg 15, ln 5, and Figure 2.

Referee Comment:
Below approx. 2 µm no modeled 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)
Author's response: This is an excellent point. Calculations for a wider range of ice crystal sizes are now included and discussed. See previous response.

Referee Comment:
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.
Author's response: Please see our response above. This section has been expanded. The differences at larger diameters are mentioned in the text, although there is not a theoretical explanation for the observed differences.

Referee Comment:
What are the uncertainties associated with the modeled results. Errors bars should be included for the individual data points to render a comparison possible at all.

Author's response: Errors bars are not available. This is a tricky question. In the case of the modeling, the model is highly accurate for the chosen inputs. The uncertainty arises from assignments of the correct inputs. In this case, by far the largest uncertainty in the modeled results in the choice of shape. This is way we include 3 shapes. Other inputs, including wavelength and particle diameter, refractive index are known with high precision.
Authors' changes in the manuscript: The uncertainty which arises due to particle shape is now explicitly states, "It is not known which of these habits best represents individual ice crystals nucleated and grown in the CFDC. Fortunately, if it is assumed that only particles of 2 µm diameter or larger are ice crystals in the CFDC, these theoretical results shown that all water and ice particles on any of the three habits will be accurately identified."

Referee Comment: P. 14, l. 1: Please insert: “aerosol particles”
Author's response: Since "aerosols" is acceptable grammar, we prefer to keep it. This is unchanged.

Referee Comment: P. 14, l. 2: Please insert: “… shown in Fig. 1 …”
Author's response: Done.

Referee Comment:
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 …”
Author's response: We consider the original text to be more succinct in this case.

Referee Comment: P. 14, l. 5: Please change to: “… in Fig. 1a …”
Author's response: Done.

Referee Comment:
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).

Author's response: It can be seen qualitative in the figure that droplets have depolarization ratios up to 0.3. At this point in the manuscript, this is only a simple choice based on visual observation. However, in Section 3.7, optimization of the depolarization threshold is performed using linear regression analysis, and the results come to the same conclusion, that 0.3 is the preferred choice of nominal threshold.
Author's changes in manuscript: We have added text (pg 15 ln 20), that states "It can be seen from this figure that droplets have depolarization ratios up to 0.3. Therefore, we visually assign 0.3 as the nominal depolarization threshold cut-off for differentiating between ice crystals and non-ice particles. The choice on 0.3 is further evaluated in Section 3.7 below."

Referee Comment: 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?
Author's response: Fig. 1c is not discussed at this point in the manuscript, so we are unsure of the Referee's intended question.

Referee Comment:
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.
Author's response: Yes, that's correct.
Authors' changes in manuscript: Changed "the size mode" to "aerosol size distribution."

Referee Comment: P. 15, l. 13- 19: In section 3.3 you discuss the usage of a depolarization Threshold of 0.3 to discriminate between different particles 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 sub saturated, to avoid any confusion with your threshold of 0.3 discussed earlier.
Author's response: Actually, the value here is the mean depolarization reported and it is consistent with the mean depolarization of training data ice crystals. As indicated on the y axis label, Figure 4b shows the mean depolarization ratio of all particles above 2 microns at that time. Because we only consider those particles larger than 2 microns and we are not in WDBT conditions until 11:55, these particles are ice crystals.
Authors' changes in manuscript: We have expanded the previous section that discusses mean depolarization ratio of the training datasets to reduce confusion.

Referee Comment: 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.
Author's response: See previous comment.

Referee Comment:
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. Further, the two Snomax®

Cases presented are not labeled in the figure, such that the reader cannot assign a CFDC c enter temperature difference between the runs from the lines in Fig. 5.
Author's response: We agree this should be clearer.
Authors' changes in manuscript: The text is now modified. In addition, we have added labeled, "Snowmax 21 ^{o}C, and Snowmax 33 ^{o}C to figure 5.

Referee Comment: 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 error bars associated with the modeled results. Consider using standard error of the mean for normalization to number of observed particles at the different sizes.
Author's response: Please see our responses above regarding the challenges of reporting error for the modeling results.

Referee Comment:
P. 16, l. 28: Please add for clarification: "… from all FIN02 experiments and not only the Snomax® experiments discussed in Section 3.5."
Author's response: Done.

Referee Comment: P. 17, l. 1: Remove "u" after 2.
Author's response: Done.

Referee Comment: P. 17, l. 1-4: Consider deletion as you already reference to the description given in section 3.5.
Author's response: Although this is somewhat redundant, we feel it best to restate these rules to avoid any confusion.

Referee Comment: P. 17, l. 510: 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"?
Author's response: Unfortunately, no. We see a wide variety in backscatter and depolarization ratios and don't have any way to answer assess this. As an aside, we have tried collecting ice crystals exiting the CFDC in plastic casts (made from dissolving plastic in dichloroethane), but our attempts so far data were not of high enough quality to determine ice crystal habit.

Referee Comment: P. 17, l. 14: Please replace "region" by "population".
Author's response: Done.

Referee Comment: 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 other 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?
Author's response: Please look again at the figure 5 and the related discussion. The manuscript states the opposite of what the referee has said. As stated "13.5 % of ice crystals in the CFDC achieve a depolarization ratio > 0.3, compared to 1.5 % percent of water droplets and 0.3 % of

aerosols. Additionally, please note the figure 6 is showing mean values depolarization ratio. Since many of the particles detected have relatively low depolarization ratios (see figure 5 and figure 3b), this value will be low. We've added the mean throughout the discussion of figure 6 to clarify this.
Author's changes in the manuscript: "In this section, modeled and observed particles discussed in the preceding results section are compared. Fig. 6 shows modeled and observed mean depolarization ratios of particles…" "In Fig. 6, both the model calculations and the observed results indicate that ice crystals have higher mean depolarization ratios…"

Referee Comment: 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 under estimation of depolarization by CASPOL not preliminary affect smaller sized particles (that scatter in relatively less light)? Thus the discrepancies between modeled and observed results should decrease as a function of size, as the detection by CASPOL becomes more reliable?
Author's response: In general, particles scatter relatively little perpendicularly polarized light in the backward 1 raw count which translates roughly a scattering cross section of ~1 $x10^{-13}$ cm^2. This limit results in the CASPOL registering a perpendicular signal below the CASPOL's detection limit for 45 % of training ice crystals, 76 % of training aerosols, and 57 % of training droplets. In the training data sets, all particles with undetected perpendicularly polarized detector were assigned depolarization ratio of zero.
Author's change in the manuscript: The full explanation above, "In general, particles scatter relatively little perpendicularly polarized light in the backward direction, …" is now added to the text on pg 20 ln 12.

Referee Comment: P. 18, l. 22-24: How does this statement fit to your data shown in Fig. 6 (cf. "CFDC ice crystals")?
Authors' response: This statement cannot be directly applied to figure 6 since the figure shows a mean and error bars that report the standard deviation. Since only 13.5 % of ice crystals achieve a depolarization ratio of 0.3 or greater, the error bars here will not show this range of particles. Since we have focused heavily on this point in the depolarization ratio distributions previously in the manuscript, we do not wish to expand anymore here. Rather, the point of this figure is to compare the mean observed depolarization ratios to modeled depolarization ratios. No change has been made.

Referee Comment: P. 18, l. 25: This is contradictory to the values you state on p. 16, l. 8-9. Please clarify.
Authors' response: The referee is right. This was a typo in the original statement here.
Author's change in the manuscript: The text (pg 21, ln 1) now reads, "A depolarization ratio threshold of 0.3 is a favorable criterion to detect ice crystals because < 2% water droplets and aerosols achieve this depolarization ratio."

Referee Comment: P. 18, l. 27: Please clarify what signal to noise ratio you refer to.
Authors' response: The signal is ice crystals, the noise is water droplets with a depolarization ratio of 0.3 or greater.

Author's change in the manuscript: Pg 21, ln 5: "…effectively reducing the signal (ice crystals) to noise (water droplets with $\delta \geq 0.3$) ratio ~1:1 or worse."

Referee Comment: 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.
Authors' response: Agreed.
Author's change in the manuscript: The text has been modified and the depolarization ratio threshold is not referred to as optimal until after the linear regression fit has been introduced on pg. 22 ln 20. "Figure 7 shows that the 0.35 threshold out performs all other thresholds when M > 20. The mean R^2 value for the 0.35 threshold is 0.46. The next best performing threshold is 0.3 with a mean R^2 value of 0.44. However, aerosol and water droplet concentrations in CFDC experiments are typically in the range $1< M< 20$ so it is appropriate to give more weight to the performance of the fit at these values. The mean R^2 value in this range of M for the 0.3 and 0.35 thresholds 0.71 and 0.7 respectively. While the performance of these thresholds perform comparably over this range, we selected the 0.3 threshold because it will slightly outperform the 0.35 threshold, especially when detecting lower INP concentrations."

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.
Authors' response: The concentration of aerosols and droplets can change in the CFDC. This purpose of this exercise is to understand how that fit will perform over all ranges of *M*. This is stated in the manuscript where we say, "Only one fit is determined for each threshold because we cannot feasibly design a model that adapts to water droplet and aerosol concentration in the CFDC."
Author's changes in the manuscript: pg 21, ln 26: 1) "The upper range of *M* values here represents an extreme sampling condition where there are many aerosols and many CCN that will form cloud droplets, but not many INP that will form ice crystals. Given the relatively high number of aerosols and droplets, this would represent the most challenging sampling scenario for proposed new method."

Referee Comment: P. 20, l. 4: Please replace "The Fig." by "It".
Authors' response: This sentence was removed during revision of the section.

Referee Comment: 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
Authors' response: This is a good point. Please see our response above. We have added additional values and discussion.

Referee Comment: 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 "parameterization" for INP at this stage.

Authors' response: In the CFDC, we assume a one to one relationship between ice crystals and INP. There is no shattering or multiplication, so this is an accurate assumption. We believe the reviewer is alluding to field observations of ice crystals which have been larger than concurrent INP concentrations. It is far beyond the scope of this manuscript to deal with disagreements between instruments in the literature, and most importantly, that question is not applicable to the internal chamber of the CFDC.

Referee Comment: P. 20, l. 22: Please add: "Each relative humidity scan..."
Authors' response: Done.

Referee Comment: P. 20, l. 23: Please replace: "Supersaturation" by "Saturation ratio"
Authors' response: Please see above. We have chosen to keep "supersaturation" as the metric of interest throughout this manuscript.

Referee Comment: P. 20, l. 25: Before this statement the meaning of the circles and the asterisks needs to be introduced in the text.
Authors' response: Done.
Author's change in the manuscript: In section 3.8, we've modified to, "The reported concentrations reveal that the traditional (circles) and depolarization ratio (*) methods generally agree during "ice only" periods (blue symbols in Fig. 8)."

Referee Comment: P. 21, l. 11: Please specify what the value of the CASPOL uncertainty refers to. Is this the depolarization ratio signal?
Authors' response: This is the CFDC-CASPOL uncertainty in INP concentration, based on combined instrumental uncertainties.
Authors' changes in the manuscript: This statement is now included.

Referee Comment: P. 21, l. 23: Please quantify the detection limit of CASPOL or give an appropriate reference.
Authors' response: As discussed in the experimental section in considerable detail, the CASPOL is a single particle 60 Hz instrument. Please see Glen and Brooks 2013 and 2014 for characterization of instrument performance.

Referee Comment: P. 21, l. 22: Please replace "polluting" to "biasing"
Authors' response: Revised to " large water droplets being miscounted as INP"

Referee Comment: P. 22, l. 2: Please add: "...mean percent error (MPE)..."
Authors' response: Done.

Referee Comment: 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.
Authors' response: Since accuracy as a function of concentration was just discussed in detail in the previous paragraph of the manuscript, we do not wish to repeat those details here.

Referee Comment: 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.
Authors' response: Please see the experimental section in which a detailed description of the differences between the two CFDC are discussed. Most importantly, because the CSU CFDC doesn't experience WDBT until a higher RH, this comparison provides a means to evaluate performance of the new method under conditions which our traditional method is clearly failing. Also, in general, because the two CFDCs are quite different instruments, an intercomparison is worthwhile.

Authors' changes in the manuscript. For emphasis we include the following statement on page 24, ln 20 Thus, inclusion of the CSU data provides a test of the new method at higher relative humidities under conditions when data obtained through the TAMU CFDC's traditional method is clearly spurious due to water droplet breakthrough."

Referee Comment: P. 22, l. 16 – P. 23, l. 14: Is your new method not applicable to other CFDCs operated along with CASPOL at all?
Authors' response: In theory the method is application. No one has tried that yet, to the best of our knowledge.

As the comparison to the modeled results indicates, having the CASPOL's unique particle-by-particle measurements of both depolarization and size is a clear advantage for reliable particle discrimination. Depolarization alone can be used to differentiate between droplets and ice crystals. However, to differentiate between dust aerosol and ice crystals are both depolarizing, so the size information provided by the forward scattering detector is needed as well as depolarization.

Figures:
Referee Comment: 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.
Authors' response: Done.

Referee Comment: Subpanels (a/d), (b/e) and (c/f) are plotted for the same datasets. However, the color bars 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.
Authors' response: The color scales for the plots have been carefully selected for readability of the plots. The objective of the plot is to reports patterns in the optical signatures and not to compare them, so it's appropriate that the scales are different in this case. No change has been made.

Referee Comment: 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.
Authors' response: The x-axis is already plotted as a log scale. Larger aerosols and smaller ice crystals have now been incorporated into the figure, and “model” has been removed from the legend. The comma has been added to the caption.

Referee Comment: 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.
Authors' response: No other instrument was used to size particles here.
Authors' changes in the manuscript: To figure 3, we have added “as detected by CASPOL” in the caption, and added d.r. symbol to x-axis of (b).

Referee Comment: 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.
Authors' response: The large particles here are ice crystals. We’ve added a size distribution to the supplemental section that shows that there are no Snomax aerosols that are larger than 2-micron diameter.
Author's changes to the manuscript: The text now includes the supplement figure, Fig S1. and a statement referring to it (pg 17, ln 6.)

Referee Comment: 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.

Authors' response: This is a good advice. However, there are several challenges presented by the data that inhibit us from displaying the data in this manner. Because the data is not collected at regular intervals of super saturation, there would be breaks in the data that make the plots hard to decipher and likely confusing to the reader. After attempting to plot the data this way, we decided that it would be better to display the data as we have here.

Referee Comment: 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.
Authors' response: In the original version of figure 4, there was already a label to describe when WDBT happens.
Authors' response: We have added a label for “Normal Operating Conditions” to Fig. 4.

Referee Comment: Please make axis ticks more visible (e.g. reduce thickness of axes) and add ticks to x-axes in Fig 4a/b.
Authors' response: Done.

Referee Comment: Add explanation for the horizontal dashed lines in the figure caption (see p. 7, l.20).
Authors' response: Done.

Referee Comment: Caption for panel c should include the case number and a reference to Table 1.
Authors' response: Done.

Referee Comment: 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)
Authors' response: The author's are confused about what this comment is requesting since all standard deviations are reported. Additionally, we have confirmed that the standard deviations reported are correct. The x-axis is already reported as a log-scale.

Referee Comment Please change the label of the y-axis as the data is a mixture of modeled and observed depolarization ratios.
Authors' response: Done.

Referee Comment: Figure 10:
The x-axis label should read "traditional concentration".
Authors' response: Done.