

## Responses to Anonymous Referee #3

### Introduction

Thank you very much for your thorough review of our manuscript. Your comments were very helpful in improving the manuscript. We sincerely appreciate the detailed and organized comments and have incorporated them to the best of our ability. As recommended, we have tightened up the text and made critical updates to the figures for clarity. Below, we have included responses to each of your comments.

### Major Comments

1. Move Section 3.2, which describes the start-up procedure, to the Supplemental Materials. Very few readers will be interested in the details of the order in which flows are started.  
**This description has been moved to the Supplemental Materials section.**
2. Lines 172-175. Please describe these optical particle counters in more detail. Are they used to count particles or to measure size distributions? What is their size resolution (e.g., bins per decade of particle size). Are they white-light or monochromatic? Do they suffer from sizing ambiguities due to Mie oscillations? What are their flow rates and uncertainties?  
**Changes have been made to the manuscript as suggested. Please see lines 217-222. For further details please refer to the manufacturer's website (<http://www.alphasense.com/index.php/products/optical-particle-counter/>) and (<https://www.tsi.com/optical-particle-sizer-3330/>)**
3. Line 177. Please describe the PROTeGE wet particle generation system in one sentence—what does it do, and over what size range? Does it produce a monodisperse or polydisperse aerosol?  
**Additional information has been provided in the manuscript. Please see lines 145-148. Please see also the published version of the PROTeGE paper in AMTD for further details.**
4. Lines 200-201. Is this shift in residual size from 3.7  $\mu\text{m}$  dry size to a 4.7  $\mu\text{m}$  droplet residual a result of incomplete evaporation, or is it just the sizing resolution of the OPS? We don't know enough about the OPS response to evaluate whether this discrepancy is significant (see comment (2) above).  
**We cannot rule out that the particles are not fully effloresced by the time they reach the OPS which is also stated in Line 322 “that this goes along with incomplete efflorescence in the system”. Also the size bin resolution of the OPS in that range is from 3.34 to 4.16  $\mu\text{m}$  (Midpoint Diameter 3.74  $\mu\text{m}$ ) followed by the next bin from 4.16 to 5.18  $\mu\text{m}$  (Midpoint Diameter 4.67  $\mu\text{m}$ ). So if droplet residual was close to 4.2  $\mu\text{m}$  in diameter which is close to the calculation of 3.7  $\mu\text{m}$  it would still have been detected in the 4.7  $\mu\text{m}$  bin leading to this discrepancy of 1.0  $\mu\text{m}$ .**
5. Line 203. When the pumping flows are turned on, shouldn't the residence time in the sample flow increase because much of the flow is being pumped away? Wouldn't this lead to better evaporation of the particles and a size closer to the expected 3.7  $\mu\text{m}$  than the test without the pumping flows (and thus a shorter residence time)?

We believe that the referee is incorrect since the sample flow of a PCVI is only driven by the add flow and is therefore independent from the pump flow. Also the sample flow of 2 lpm when the other flows of the L-PCVI were off (1 lpm drawn from the OPS and 1 lpm via Massflow controller in total 2 lpm, see line 320) and the sample flow of 2 lpm when the flows of the L-PCVI were on, are identical so the residence times of the droplets in the sample flow are equal.

6. Table 1 has only one row, and this information could probably go into a single sentence in the manuscript.  
**We agree with your response and have adjusted the paper accordingly. Table 1 has been replaced by the in-line discussion of flows in Section 2.**
7. There are 17 figures, which is just too many for a relatively concise technical manuscript. Several figures contain peripheral information that could be moved to the Supplemental Materials without any loss of content for the vast majority of readers. These figures include Figs. 3, 4, 8, and 15. Figure 10 and the associated discussion in the text could be eliminated entirely, unless the authors provide a compelling reason to keep it. It distracts from the main thrust of the manuscript. Similarly, Fig. 13, which introduces data from an SMPS (which is not described in any way in the manuscript) doesn't really seem relevant to the topic of the paper. Presumably the SMPS is a scanning mobility particle sizer, which is generally heavily weighted toward particles too small to serve as IN in most environments, and so is really a tangent to the main topics of the manuscript.  
**Figures 3, 4, 8, 10, 11, and 13 were moved to the supplementary section and additional sentences were provided to help explain this. Figure 10 provides additional information about the particle distribution in the PF, which is important for SPIDER's operation. The purpose of Figure 13 was to help demonstrate that the aerosol particles in the ambient environment were low and would minimally contaminate ice crystals.**
8. Figure 16 is just a blow-up of a section of Fig. 14. I recommend that these figures be combined, with the shading added to Fig. 14 and Fig. 16 eliminated.  
**Figure 16 was removed. Figure 14 was updated with shading.**
9. The caption for Fig. 14 is for a different figure. The top row of Fig. 14 appears to be data from a cloud probe, and the lowest row to be data from the L-PCVI (which also appears in Fig. 16). I have no idea what the middle row is. Some legends would be helpful for quickly reading the figure. As I mentioned, I don't see a need to replicate the data in Fig. 16; the data are pretty clear in Fig. 14.  
**Figure 14 was removed.**
10. All of the size distribution figures (e.g., Figs 6, 9, 10 (if kept)) and 17 should be converted to follow the dN/dlogD<sub>p</sub> convention. They are currently histograms that are not normalized by bin width. Thus any peaks in the distribution could just be caused by a wider diameter bin than other bins in the distribution, rather than an actual peak in the probability distribution function. It's too much to expect the reader to look at the diameters labeled on the axis (in a tiny font) and try to figure out if all the bins are the same logarithmic increment or not. A more traditional PDF display using dN/dlogD<sub>p</sub> with a log x-axis would be much appreciated.  
**All of the aforementioned figures have been updated to dN/dlogD<sub>p</sub> convention.**  
**Additionally, fonts have been increased to increase readability.**
11. On many of the figures, the font weight is too light to be quickly and easily read. In Figures 3, 4, and 8 (hopefully moved to SI), the curves are hard to distinguish. Please use a heavier line weight

and colors (preferably using a palette that is visible by the color-impaired) to help distinguish them.

**Font changed to Arial and darker colors were used.**

12. In Figure 5, please describe the meaning of the "X", arrow, and circle marks on the icons for the PCVI. This is a really clever idea to show how the sampling was set up; just describe it the first time it's used, please.

**We have added text to Section 4.1 and the figure caption to provide additional context for this figure.**

13. Figure 11 is great, but the symbols need error bars.

**Representative error bars have been added to a point near the D50 and to the D50 itself.**

14. Figure 15 could be removed; just state that the ice crystal residual concentration was not correlated with wind speed.

**Figure 15 was moved to the supplementary section of the document since there was no correlation.**

15. In Figure 7, I don't see any distinction between "dark grey" and "light grey" bars. The text needs to more clearly describe what these bars are and how they were calculated. The rightmost bar appears to go off the top of the figure; it would be interesting to know how far up it goes. If the bars represent the range of values, there is really a LOT of spread to this measurement and it is really not too useful for interpreting performance. It could be moved to Supplemental Materials. Perhaps a box-and-whiskers presentation would be more useful than the bars?

**Figure 7 was updated based on the comment. Edits include simplifying the legend, darkening the color scheme, and increasing the y-axis bounds to show that the upper bound of the range is 50 um. This figure is also moved to the Supplementary Materials section.**

## Minor Edits

- a. Line 78: "design CVI"

**Change made.**

- b. Line 123: Is there any risk of nucleating ice particles (heterogeneously) in the chamber at -16 C?

**Author Response: All flows entering the system are fully filtered during laboratory and field studies. In the literature is known that there is some inadvertent transmission of particles ~ 10-100 particles/Liter but a publication from Demott et al. showed that only 1 particle/1 million particles acts as in INP. Therefore we would need to run ~10000 liter of air through the chamber to get to 1 million particles to make this a value to be important.**

- c. Line 126: Changed to "machined PCVI". It's just awkward as written, even if grammatically correct.

**Change made.**

- d. Line 155: change to "containing the same wavelength as the printer's laser"

**Change made.**

- e. Change to "Figures 5a and 5b show"

**Change made.**

- f. Lines 230-232. The first sentence says that droplets from 25-50  $\mu\text{m}$  evaporate, while the second says that droplets larger than 25  $\mu\text{m}$  will not evaporate. Please reconcile  
**Change made.**
- g. Line 236. Change "in the  $\sim 20 \mu\text{m}$ " to "of the order of  $\sim 20 \mu\text{m}$ ".  
**Change made.**
- h. Line 241. What does "close to the entry point of the chiller" mean. Does this mean that the droplets had to pass through extremely cold air on the way to the chamber entrance? This is rather unclear. This is not shown on the schematic diagram of the apparatus (Fig. 2).  
**Updated text and added a marker to Figure 2 to show where the chiller fluid enters and exits the chamber.**
- i. Line 242. Change to "would rapidly freeze".  
**Change made.**
- j. Line 254. This description is not consistent with Fig. 10b.  
**Updated inline.**
- k. Line 287. Change to "set up"  
**Change made.**
- l. Line 288: Change to "scattering signal from the Droplet Measurement Technologies. . ." Readers may wonder why a BC detector is used for non-BC particle concentration measurements.  
**Change made.**
- m. Starting with line 304, there is a lot of discussion of the minutiae of blowing snow, etc. This section of the manuscript is not very relevant to the analysis and could be condensed and the figures moved to Supplemental Materials.  
**Deleted second sentence beginning in line 304 continuing to line 307. Only a brief comment about blowing snow not influencing ice crystal residual concentrations is left. The figure was moved to the supplementary section.**
- n. Line 317. Change to "in the Aitken mode to fine particles in the accumulation mode".  
**Change made.**
- o. Line 318: What is meant by "where coagulation and condensation are taking place"? There is little coagulation between accumulation mode particles. I'm not sure what the intent of this sentence is.  
**Sentence was deleted for clarification.**
- p. Line 318. Define what is meant by the "ice crystal channel".  
**The first paragraph was updated to define what the ice crystal residual channel meant. "The scattering signal from the Droplet Measurement Technologies (DMT) Single Particle Soot Photometer Extended Range (SP2-XR) was used to measure ice crystal residuals. This was located downstream of the SF of the PCVI and is called the ice crystal residual channel."**
- q. Line 321: Define what is meant by the "cloud droplet residual channel".

**The first paragraph mentions was updated to define what the cloud droplet residual channel meant as well as the interstitial aerosol channel. “Two OPCs were attached to SPIDER and were used to measure interstitial aerosols and cloud droplet residuals. The PF from the L-PCVI is where the OPC counted the interstitial aerosols and is termed the interstitial aerosol channel. The PF of the PCVI is where another OPC was located and counted the cloud droplet residuals and is called the cloud droplet residual channel.”**

- r. Fig.1 caption. Suggest replacing "slanted" with more technical description. Are you discussing the angle of the conical inlet? I would describe it as "more tapered".  
**Updated to “conical”**
- s. Fig. 2. What are TS01-TS16 shown in the figure? I assume these are temperature sensors but they are not mentioned in the text.  
**The figure has been updated to define the acronym explicitly. Additionally, text has been added to explain what these are.**