Response to Anonymous Referee #1

The authors wish to thank Referee #1 for his/her thoughtful comments and useful discussions. Below are our point-by-point responses (in blue texts) to the reviewer's comments. Corresponding modifications are reflected in the manuscript and figures.

Major comments:

Reviewer's comment: 1) Section 4.3, Transmission efficiency. It was not clear how Figure 7 was generated. I went back and read the TE analysis and the methodology in Gallavardin et al. 2008. From the description in Gallavardin et al. 2008 it was clear how the NOAA PCVI transmission curve was generated, but I was still confused on how the open symbols were generated for the IS-PCVI in Figure 7. In addition the brief description in section 4.3 on how the transmission efficiency was determined seems to be different from what is in Gallavardin et al. 2008. The description of how the transmission efficiency was determined needs to be expanded and improved in the current manuscript for clarity.

Authors' response: The reviewer is right. The method for our TE estimation is slightly different from that of *Gallavardin et al.* (2008, AS&T, G08 hereafter). For instance, we estimated the TE curve by comparing residue concentrations measured by the CPC 2 downstream of the PCVIs to activated particle concentrations (= the sum of particles above D_c, which is 15.3 ± 1.9 µm) estimated by the AIDA OPCs by following the method described in P776 of G08. The only difference between G08 and the current study is the derivation of the droplet diameter (i.e., the x-axis of Fig. 3a in G08). While G08 used the FTIR to determine the liquid droplet diameter over the course of the expansion, we used the count median diameters of the AIDA OPCs (5 seconds time resolution). To improve the clarity and address the reviewer's question, we have modified P14 L16-20 as follows:

"To be comparable to the previous AIDA study, we used a somewhat similar methodology of the TE evaluation described in *Gallavardin et al*. (2008). Briefly, the TE spectrum of the IS-PCVI was understood by plotting the ratio of droplet residual concentration measured by the CPC 2 after the IS-PCVI to the activated droplet number concentration above its D_c (15.3 ± 1.9 µm; Table S2) as function droplet diameters in D_{ve}. The only difference between *Gallavardin et al*. (2008) and the current study is the derivation of the droplet diameter. While *Gallavardin et al*. (2008) used Fourier transform infrared spectroscopy to determine the liquid droplet diameter over the course of the expansion, we used the count median diameters of the AIDA OPCs (i.e., OPC 1 and 2). Despite the difference, we successfully generated supercooled droplets up to ~25 µm D_{ve} in the chamber. The IS-PCVI was operated with 7 lpm CF and 70 lpm IF.".

Reviewer's comment: 2) Also related to the above, in Section 4.3 Dc was 15.3 micrometers but the estimated 50% cut-size for the particular flow conditions was 9 micrometers based on Figure 7. Shouldn't these numbers be the same? I.e. shouldn't Dc equal the 50% cut-size?

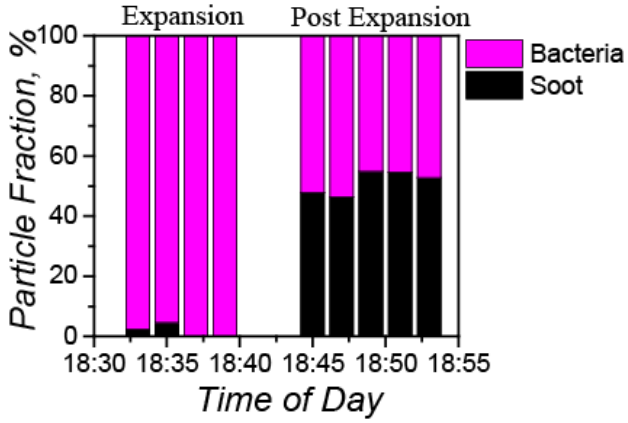
Authors' response: D_c ~15 µm and D_{50} ~9 µm are correct interpretations. As prescribed in Sect. 3.2.3, D_c ("critical" cut-size) is optimized as a diameter equivalent to the mid-bin size of the point where INPs and/or CCNs ≈ residuals. Therefore, D_c is not necessarily equal to D_{50}. In other words, D_c can be larger than D_{50}.

Reviewer's comment: 3) Page 17, line 16. ". . .with only a small amount of soot (<5%) making it through the IS-PCVI". This assumes that the minisplat has the same sensitivity to soot as bacteria. My understanding is that single particle mass spectrometers may not detect small particles, such as soot, with 100% efficiency. The authors should discuss the detection efficiency of the minisplat to soot and bacteria as a function of size and discuss the implications for their findings.

Authors' response: The instrument's detection efficiency as a function of particle size was discussed in a previous publication (*Zelenyuk et al*., 2015), referenced in the present manuscript. To clarify this point, we added the following sentence:

P12 L4: The detailed discussion regarding the instrument's performance (e.g., detection efficiency as a function of particle size) is available in *Zelenyuk et al*. (2015).

The relative soot/bacteria particle number concentrations reported here as well as absolute numbers obtained through other means are correct as they are. To verify the capability of miniSPLAT's bacteria/soot detection downstream of AIDA and IS-PCVI, we measured the relative soot/bacteria ratio without counterflow after the expansion (i.e., FIN01_38). The figure below shows that miniSPLAT successfully measured approximately 50:50 of soot and bacteria particles in the AIDA chamber (see Post Expansion time-resolved columns), supporting that the substantially low soot fraction during the expansion, which is <5% (and below detection later), is due to inertial separation but not because of the instrument's detection issue.



Minor comments:

Reviewer's comment: 4) Page 5, line 6. "more specifically the 5 ohm heating wire is varied inside the heat conductive nozzle tip around the converging section over the 35 mm length, keeping the . . ." Is the heating wire varied or is the current through the heating wire varied?

Authors' response: The latter is correct. For clarity, we rephrased P5 L6-7 to "…the 5 Ω heating wire is installed inside the heat conductive nozzle tip around the converging section over the 35 mm length. The current through the heating wire is varied depending on the surrounding gas temperature (*T*) to keep the nozzle surface *T* above the sampled air *T* to prevent frost formation and growth."

Reviewer's comment: 5) Page 8, line 20. "ddH2O". Typo?

Authors' response: Thank you. Corrected (ddH_2O = double-distilled water).

Reviewer's comment: 6) In a few equations, symbols are not fully defined. For example, what is V(infinity) and V(vapor) in equation 8. The meaning of the symbols can be figured out from the context, but it would be clearer to define all symbols.

Authors' response: The reviewer makes a good point. Now P13 L1- reads as follows:

"Conceptually, this factor can be estimated by relating the measured time rate change of diffusional growth of ice mass inferred by the OPC measurement ($\frac{dM}{dt}_{OPC}$) to the predicted rate ($\frac{dM}{dt}_{predicted}$; *Hinds*, 1999):

$$\frac{dM}{dt}_{predicted} = \frac{dM}{dt}_{OPC} \times \Psi, \qquad [7]$$

Alternatively, we can also optimize this factor by relating the total condensed water volume ($V_{optimized}$), which can be estimated by subtracting the vapor phase water volume (V_{vapor}) inferred by in situ by tunable diode laser water vapor absorption spectroscopy (*Fahey et al.*, 2014) from the total water content in the chamber (V_∞) inferred by a fast high precision chilled mirror hygrometer (MBW, model 373; *Wagner et al.*, 2008), to that derived from OPC (V_{OPC}):

$$V_{optimized} = V_{\infty} - V_{vapor} = V_{OPC} \times \Psi. \quad [8]"$$

P_{CVI} is now defined in Sect. 2.4 as, "The IS-PCVI *P* (P_{CVI}) was monitored…".

In Sect. 3.3, we define $N_{residual}$ and $N_{CPC\ 2}$ as, "To obtain the number concentrations of the IS-PCVI residuals that are comparable with the AIDA droplet and ice particle size distributions ($N_{residual}$), the number concentration of residuals measured downstream of the IS-PCVI ($N_{CPC\ 2}$) has to be corrected for **i)** the calibration factor of the CPCs and **ii)** the particle concentration factor:". In addition, $\frac{CPC_{upstream}}{CPC_{downstream}}$ is defined in Sect. 3.3 as, "the calibration factor of the CPCs ($\frac{CPC_{upstream}}{CPC_{downstream}} = \frac{CPC\ 1}{CPC\ 2}$) is…".

Added Ref.: Fahey, D. W., Gao, R.-S., Möhler, O., Saathoff, H., Schiller, C., Ebert, V., Krämer, M., Peter, T., Amarouche, N., Avallone, L. M., Bauer, R., Bozóki, Z., Christensen, L. E., Davis, S. M., Durry, G., Dyroff, C., Herman, R. L., Hunsmann, S., Khaykin, S. M., Mackrodt, P., Meyer, J., Smith, J. B., Spelten, N., Troy, R. F., Vömel, H., Wagner, S., and Wienhold, F. G.: The AquaVIT-1 intercomparison of atmospheric water vapor measurement techniques, Atmos. Meas. Tech., 7, 3177-3213, doi:10.5194/amt-7-3177-2014, 2014.

Reviewer's comment: 7) Page 14, lines 25 to 30. "The outline heating was turned off to prevent evaporation of droplets." Can there still be some evaporation due to the addition of the dry counter flow?

Authors' response: Correct. Our counterflow has *T* ~20 °C and, thus, may contribute to droplet evaporation. We calculated the droplet size reduction due to evaporation while droplets traveling from the point where counterflow (20 °C) meets with the droplet stream to the downstream OPC (~1 m below). Our estimation based on *Hinds* (1999) assuming a saturation ratio <1 shows that there could be up to 30% size reduction. The relative importance of the evaporation to the other factors mentioned in P14 L29-30 is unknown. Nevertheless, the resulting overall TE result, which is based on the measurement/observation, accounts for all factors and, therefore, does not change.

For clarity, we rephrased P14 L29-30 to, "The observed particle losses may have occurred in the external inlet and/or the IS-PCVI. Droplet evaporation due to the dry counterflow may also have played a role. Neverthless, …".

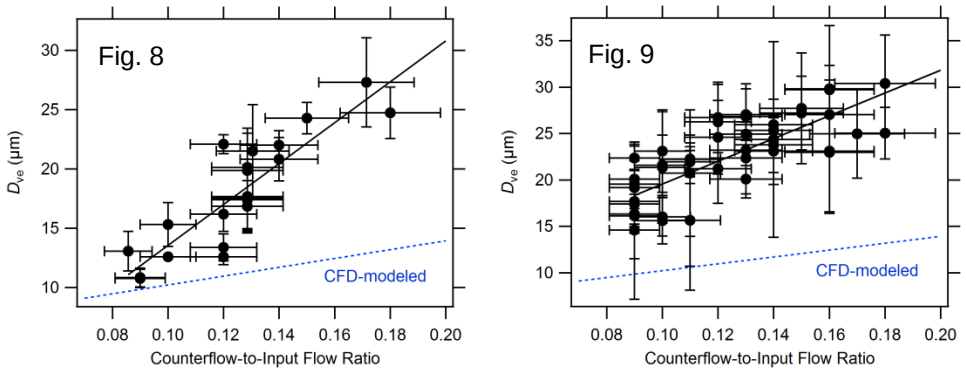
Reference: Hinds, W. C.: Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles, 2nd Edition. Wiley-Interscience, New York, NY, USA, 278–303 pp., 1999.

Reviewer's comment: 8) Page 14, line 9-11. "During this expansion, both OPC 2 (>15 microns) and CPC2 counted > 0.1 cmˆ3 of ice residuals. . .". Should this sentence be modified since OPC 2 does not count residuals, rather it counts ice crystals.

Authors' response: The reviewer is correct. Thank you very much for your suggestion. We rephrased the sentence as, "During this expansion, the sum of particle concentrations above 15 µm D_{ve} detected by OPC 2 exceeded 0.1 cm^3 after 15:26:40. Likewise, CPC 2 counted >0.1 cm^3 of ice residuals for the same period of the experiment.".

Reviewer's comment: 9) Figure 8 and 9. The trace for CFD-modeled is not a straight line?

Authors' response: The authors would like to thank the reviewer for pointing out the CFD trace behavior. Previously, we used incorrect CF to IF ratio's to plot the CFD measurements. In the revised version, we used correct ratio to plot the CFD results. The figures are revised, as shown below.



Reviewer's comment: 10) Page 15, line 8. "the linear relation does not hold if the ratio is outside these bounds". Change this to "the linear relation may not hold if the ratio is outside these bounds" unless you have data to prove otherwise.

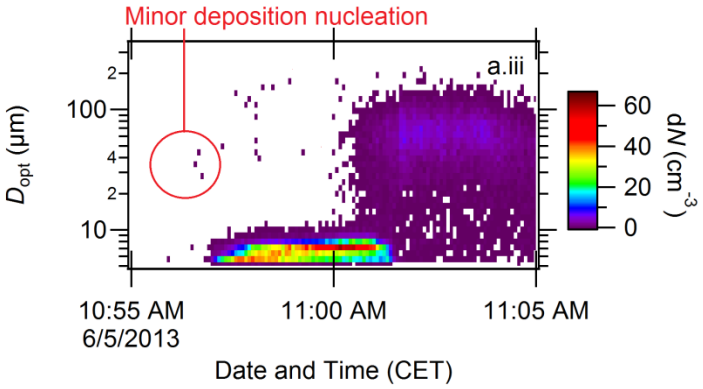
Authors' response: Changed. Thanks for the suggestion.

Reviewer's comment: 11) Page 16, line 3. "the sum of the CPC 2 and CPC 3 counts < the CPC 1 counts". From the figure it looks like the sum of the CPC 2 and CPC 3 counts are > the CPC 1 counts". For example at a CPC 1 count of 150, the CPC 2 + CPC 3 > 150. What am I missing?

Authors' response: This is a good point. To improve the clarity of this part, P16 L3-4 is now rephrased as, "Though the flow dilution or concentration factors ($\frac{F_{output}}{F_{input}}$ and $\frac{F_{input}}{F_{pump}}$ for CPC 2 and CPC 3, respectively) and the particle losses through the CPCs (i.e., ~25% for CPC 2 as abovementioned in Sect. 3.3 and ~10% for CPC 3) are accounted in our CPC counts reported in Fig. 11, the concentration accuracy of CPCs in the manufacturer's report (±10%) cannot be ruled out as a source of data diversity. Regardless, our observation suggests that the total particle number density measured by CPC 1 agrees with the sum of residual density in the sample flow (CPC 2) and interstitial aerosols in the pump flow (CPC 3) within ± 10%. We note that a maximum of 8% loss of droplets in the IS-PCVI can occur (i.e., the sum of the CPC 2 and CPC 3 counts < the CPC 1 counts).".

Reviewer's comment: 12) Page 16, line 10-11. "the minor deposition mode nucleation was also detected by the AIDA OPCs (fig 12 a.iii) around 10.56." I don't see this in the figure. Can annotation be added to point out what the authors are referring to?

Authors' response: For clarity, we modified the sentence as, "…the minor deposition mode nucleation was also observed around 10:56 (~0.12 cm^{-3}) before the droplet formation was observed by AIDA OPCs (Fig. 12a.iii).". As shown below, first three dots above 20 µm optical diameter appearing prior to the droplet formation correspond to the contribution by deposition nucleation.



Reviewer's comment: 13) Figures. In figure 11 and figure 14 has the CPC2 been corrected for the enhancement factor of the CVI (this should be made clear in the figure caption or somewhere). Also in figure 14, has the numbers for the minisplat been corrected for this enhancement factor of the CVI?

Authors' response: The reviewer is correct about CPC 2 counts, for which we report the corrected values. We clarified this point in our response to the review's comment (11). For Fig. 14, we added the following notes in the figure caption: "The residual counts measured by CPC 2 are corrected according to Eqn. 6, while no enhancement factor correction is included for miniSPLAT concentration.". We point out that we used miniSPLAT to determine the relative soot/bacteria transmission through the IS-PCVI. miniSPLAT-measured residual counts are lower when compared to uncorrected CPC 2 counts as expected due to transmission losses in lines and detection efficiency. We note a good agreement between temporal evolution of the miniSPLAT-measured total concentration of residuals and the uncorrected CPC2 counts, which are scaled here for comparison, as shown in the figure below.