Response to Anonymous Referee #2

The authors wish to thank Referee #2 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.

General Comments:

Reviewer's comment: A discussion of the paper on the "Ice Selective Inlet" by Kupiszewski et al., [2015] is glaringly missing from the introduction section. The comment on Page 4, Line 1 that "this study presents the first application of a PCVI for analyzing only pristine ice crystals from a mixture of droplets and interstitial particles" should be re-phrased in light of the Kupiszewski paper.

Authors' response: The reviewer makes a good point. We apologize for missing such an important previous work. We now discuss *Kupiszewski et al*. (2015) in Sect. 1.2. More specifically,:

P3 L10: *Kupiszewski et al*. (2015) has been added as an example of the field application of PCVI.

P3 L17- before "However, since…": We added the following sentence:

"More recently, *Kupiszewski et al*. (2015) developed a phase discriminating inlet, so called the Ice Selective Inlet, by connecting a cyclone impactor and an evaporation section upstream of a commercial off-the-shelf PCVI (BMI, Model 8100).".

Additionally, the authors removed P3 L32-P4L1 to give full credit to *Kupiszewski et al*. (2015).

Added Ref.: Kupiszewski, P., Weingartner, E., Vochezer, P., Schnaiter, M., Bigi, A., Gysel, M., Rosati, B., Toprak, E., Mertes, S., and Baltensperger, U.: The Ice Selective Inlet: a novel technique for exclusive extraction of pristine ice crystals in mixed-phase clouds, Atmos. Meas. Tech., 8, 3087-3106, doi:10.5194/amt-8-3087-2015, 2015.

Reviewer's comment: Although some descriptions of the experiments labeled by CAMPAIGN_## are described in the experiment section and Table S2, it would be useful to re-described these experiments in the results section. At the least, describing the aerosol type would help prevent the reader from having to flip back and forth between the experiments, supporting information, and results section while reading the results section.

Authors' response: P15 L12: We added "INUIT05_1-13 conducted with dry dispersed hematite particles".
P12 L7-9: One expansion experiment (FIN01_38) was designed to examine if intrusion could occur to the IS-PCVI outfit using a mixture of soot particles and the other particle type (i.e., bacteria particles, Sect. 4.8) possessing different chemical and ice-nucleating properties.

Other than those, the aerosol types are described for individual experimental IDs at their first appearance in text.

INUIT05_29 and _36: Sect. 3.2.3	INUIT05_09 and_12: Sect. 3.2.4	INUIT05_34_51_58: Sect. 3.2.5
INUIT05_55_56: Sect. 3.2.6	FIN01_04: Sect. 4.2	FIN01_38: Sect. 4.8

To avoid redundancy, we would like to keep it as is.

Reviewer's comment: This occurs specifically in sections 4.3, 4.5, 4.6, 4.7, It should be made clear throughout the manuscript when CPC 2 has been corrected using Equation 6. Furthermore, it seems to the reviewer that the "corrected" CPC 2 should always be used when determining transmission efficiencies (Section 4.3, 4.6).

Authors' response: This is a good point. We clarified this point in P12 L24 as follows:
"The residual concentrations, which are measured by the CPC 2 and reported in this study, are corrected based on Eqn. 6, unless otherwise stated.".

In general, all analyses presented in Sects. 4.3 and 4.6 have been conducted using the corrected residual concentrations. Exceptions like the measurement without any flows (e.g., Sect. 4.5) are noted: i.e., Fig. 10 caption – "The enhancement factor correction was not applied due to the static mode operation of the IS-PCVI.".

More specific corrections/modifications of texts in Sects. 4.3 and 4.6 are described below.

Reviewer's comment: Similarly, in Section 4.3, is the (OPC 3)/(OPC 1)ratio on Page 4, Line 27 also corrected for the concentration factor due to the PCVI?

Authors' response: Correct. The enhancement factor correction is accounted for the OPC 3 counts. For clarity, we now added the following sentence in P15 L10.

"The PCVI flow concentration factor correction is accounted for the OPC 3 counts.".

Reviewer's comment: Finally, in Section 4.6, is there an expected concentration factor for the pumped flow? That is, is it valid to assume that (CPC 1) = (corrected CPC 2) + (CPC 3)?

Authors' response: CPC 1 = corrected CPC 2 + corrected CPC 3 [= CPC 3 * (CPC 1/ CPC 3) * ($F_{\mathrm{Input}}/F_{\mathrm{Pump}}$)]

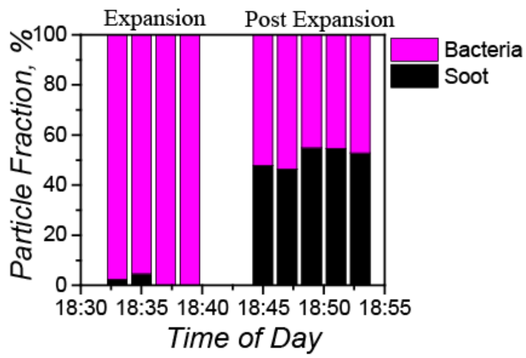
For clarity, P 16 L3-4 now reads, "Though the flow dilution or concentration factors ($\frac{F_{output}}{F_{input}}$ and $\frac{F_{input}}{F_{pump}}$ for CPC 2 and CPC 3, respectively) and the particle losses through the CPCs (i.e., ~25% for CPC 2 as abovementioned in Sect. 3.3 and ~10% for CPC 3) are 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: The reviewer was extremely happy to see the experiments in Section 4.8. It would be useful for the reader to know the efficiency of the miniSPLAT to see both the bacteria and BC. If either of those are not seen with unity efficiency, the concentrations should be corrected.

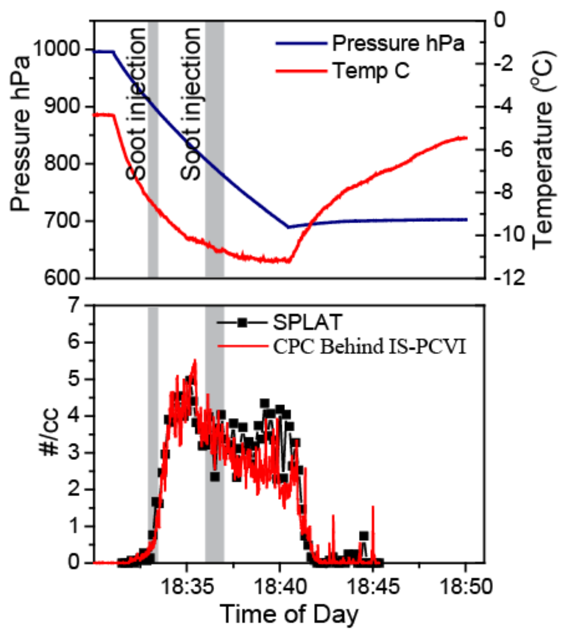
Authors' response: The instrument's detection efficiency as a function of particle size was discussed in a previous publication (*Zelenyuk et al*., 2015), referenced in the present manuscript. To clarify this point, we added the following sentence:

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 ontained 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.



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.



Reviewer's comment: Finally, can the authors postulate why the IS-PCVI seems to largely avoid the problems with wake capture, etc. that troubles the traditional PCVI as in the cited Pekour and Cziczo [2011] paper?

Authors' response: We report our artifact fraction within our experimental conditions specifically employed in this study as indicated in P18 L10-. As compared to the PCVI used in *Pekour and Cziczo* (2011), our new IS-PCVI is manufactured with a differently geometry to enable facilitating much larger counteflows and associated cut-sizes (e.g., 25 µm for FIN01_38; P17 L11). The CFD simulations implemented to guide the design prior to the fabrication process (Sects. 3.1 and 4.1) may have helped to improve the overall performance of the new IS-PCVI. Given differences in experimental setup and approach, we would like to minimize the direct comparison between two independent studies regarding the artefacts.

Minor Corrections:

Reviewer's comment: Page 2, Line 8: Are "ice residual particles" always "leftover INPs?"

Authors' response: The reviewer makes a good point. As an exception, unwanted components thorough wake capture, scavenging, collision and/or coalescence may be seen as part of ice residuals. Though these artifacts are well discussed in Sect. 1.2 and Sect. 3.2.7, we clarify this point by rephrasing this part to the following:

"The analysis of ice residual particles, which are mainly leftover INPs after evaporating the water content…".

Reviewer's comment: Page 2, Line 16: In the context of an "ice-selecting"-PCVI, it seems as if the statement "separating ice residuals from interstitial particles" would benefit from a small discussion on mixed-phase clouds and the relative size of droplets and ice particles.

Authors' response: The reviewer is right. We now rephrased this sentence to "Here, separating ice residuals from large droplets without a substantial influence of artifacts (e.g, transmission of interstitial particles) is a challenge, and ...". This modification gives a smooth link to the discussion of ice vs. large droplet given in Sect. 1.2 (i.e., P3 L24-28). We note that the size of droplets in ambient mixed-phase clouds is typically up to 30 µm diameter (above 25 µm droplets are rare) according to *Miles et al.* (2000) and *Lawson et al.* (2011). We also note that our IS-PCVI is capable of >25 µm inertial separation. For clarity, now P3 L27-28 reads "…droplets as large as a few tens of microns (< 30 µm diameter; *Miles et al*., 2000; *Lawson et al*., 2011)…".

Reviewer's comment: Page 9, Line 22: TSI 3076 is an aerosol generator, please correct. Furthermore, is this CPC what is eventually termed "CPC 1?" If CPC 1 and CPC 2 are different models, is there a possibility that you could be over/underestimating your losses through the IS-PCVI due to different counting efficiencies?

Authors' response: We thank the reviewer for catching this typo. CPC 1 and CPC 2 are identical models (i.e., 3010, P10 L3-4) and adequately calibrated (P12 L17-19).

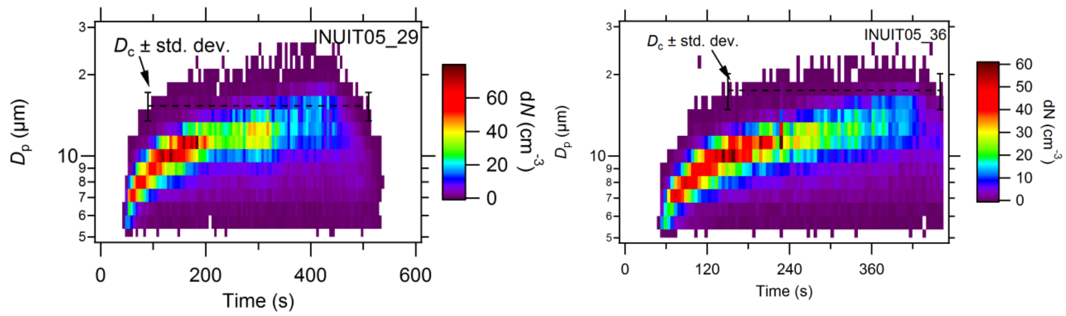
Concerning CPC 1 vs. CPC 3, P16 L3-4 is now rephrased as, "Though the flow dilution or concentration factors ($\frac{F_{output}}{F_{input}}$ and $\frac{F_{input}}{F_{pump}}$ for CPC 2 and CPC 3, respectively) and the particle losses through the CPCs (i.e., ~25% for CPC 2 as abovementioned in Sect. 3.3 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: Page 14, Line 7: A description of the dashed black line in Figure 6d should be added here. Furthermore, a discussion of why OPC 2 counts are much larger than the "corrected" CPC 2 counts should also be added.

Authors' response: We agree. We now added the following sentence, "The residual concentrations corrected based on Eqn. 6 (i.e., CPC 2 corr. in Fig. 6d) were smaller than the OPC 2 counts (i.e., >15 µm D_{ve}) all the time, suggesting that the actual cut-size of ice crystals penetrated through the IS-PCVI was much larger than 15 µm. As expected, the estimate D_c was ~24 µm D_{ve} (panel e)."

Reviewer's comment: Page 14, Line 24: It is interesting that the authors validated that the PCVI is "ice-selecting" by determining the TE of very large droplets. How prevalent would droplets of this size be for a normal AIDA chamber expansion? What about for ambient mixed phase clouds?

Authors' response: Below, we present the number size distribution of the droplets measured by OPC 2 during two relevant expansions (i.e., INUIT05_29 and 36) that are discussed within the Sect. 4.3. The time series plots of droplet size evolution are shown in the size range of 5-30 µm optical diameter. The estimated D_c ± standard deviation values (reported in Table S2) are also shown. As can be seen, the abundance of the droplet number concentrations around D_c is in the order of a few per cm^{-3}. We note that these concentrations are an order magnitude higher than our detection thresh of the welas-OPC (*Hiranuma et al*., 2014, ACP), verifying our analyses are quantitative.



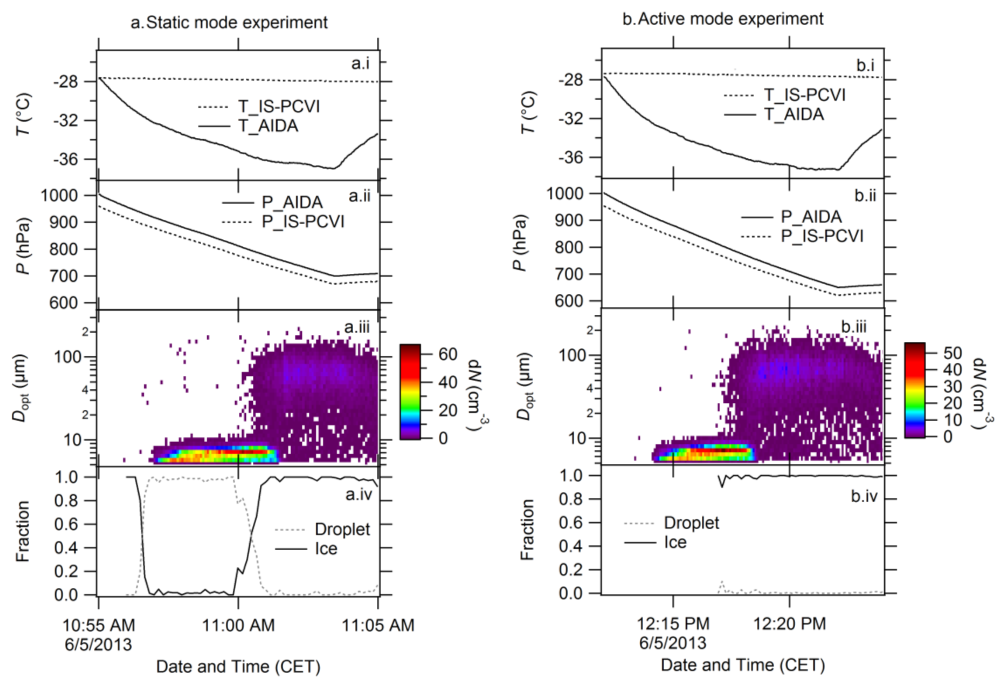
The size of droplets in ambient mixed-phase clouds is typically up to 30 µm diameter (above 25 µm droplets are rare) according to Miles 2000 and Lawson 2011 (P3 L25-28). Note that our IS-PCVI is capable of >25 µm inertial separation. For clarity, now P3 L27-28 reads as, "…droplets as large as a few tens of microns (< 30 µm diameter; *Miles et al*., 2000; *Lawson et al*., 2011)…"

Reference

Hiranuma, N. et al., Atmos. Chem. Phys., 14, 2315–2324, 2014.

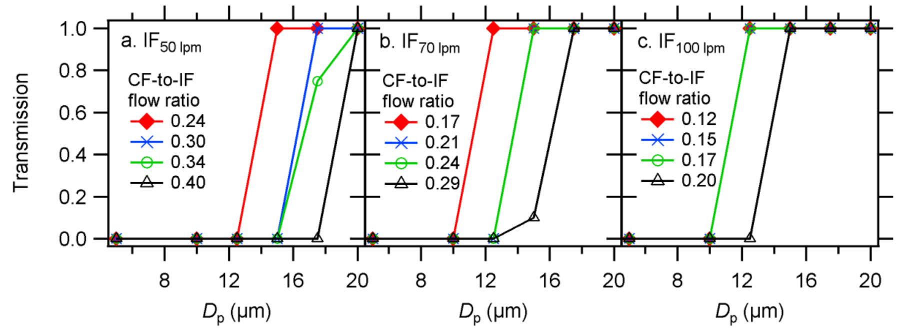
Reviewer's comment: Page 16, Line 7: It would be helpful to the reader to address the difference between 12a and 12b and define static vs. active sampling with the PVCI.

Authors' response: We added a brief description of static/active in the figure caption as, "While the IS-PCVI behaves as a regular outlet without any flows in the static mode experiment **(a)**, all flows are in during the active mode experiment **(b)**.". We also added static/active in each column of Fig. 12 for clarity.



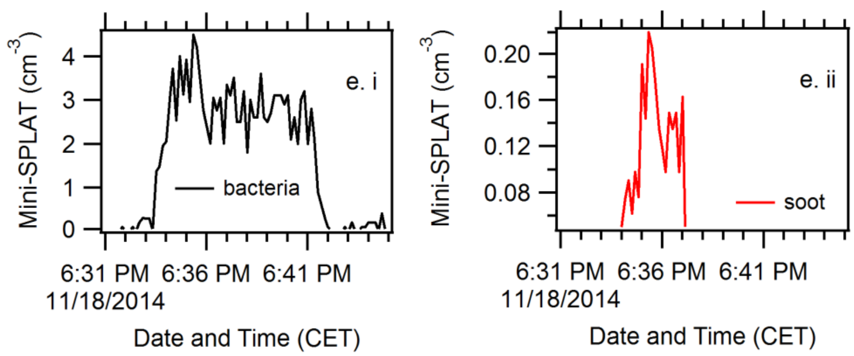
Reviewer's comment: Figure 5: It might be more instructive to the reader to have the IFs directly on each figure.

Authors' response: This is a good suggestion. Figure 5 has been modified accordingly.



Reviewer's comment: Figure 14e: It is very difficult to determine the BC concentrations after the second injection. Would it be more useful to put these concentrations on a log scale?

Authors' response: We believe that <5%, which we report in text, is a good representation of our measurements, and we would like to keep it as is. As indicated in the first page of our supplementary information (L19-21), any detailed information regarding our experimental data (i.e., concentration and size distribution of aerosol, droplet and/or ice) before and during individual AIDA expansion experiments are available upon request. The authors are willing to provide the time series plot or data (as shown below) upon individual requests. Note that no substantial BC-containing particles were measured after the second injection within our detection limit. A stable condition (small *T* reduction) inside the chamber may have helped the PCVI to perform better as compared to the abrupt cooling period.



Technical Corrections:

Page 1, Line 19: Change "in the controlled mixed-phase cloud system" to "in controlled mixed-phase cloud systems"
Changed.
Page 1, Line 21: Delete "(CFD)" as it is not used in the remainder of the abstract
Thank you. Deleted.
Page 2, Line 7: What does "their" correspond to?
For clarity, the authors rephrased the sentence to, "However, the atmospheric INP abundance and their identities as well as sources remain unanswered (e.g., *Knopf et al*., 2014).".
Page 2, Line 21: Change "to collect" to "the collection of"
Changed.
Page 3, Line 17: Delete "since"
Deleted.
Page 3, Line 20: Change "reduced the TE" to "reduced TE"
Changed.
Page 4, Line 10: Unsure what "the application" refers to

To improve the clarity, we modified the sentence as, “Further, we operated online high resolution single particle mass spectrometers behind the IS-PCVI in the *Fifth International Ice Nucleation workshop* (FIN-1) to demonstrate the capability and application of such combination for the simulated cloud residual studies.”.
Page 5, Line 5: Delete "the" in "equipped with the"
Deleted.
Page 5, Line 6: Delete "the" in "the 5-ohm heating wire"
Deleted.
Page 7, Line 7: Change "is used to seal" to "are used to seal"
Changed.
Page 8, Line 18: Change "using first two" to "using the first two"
Changed.
Page 8, Line 28: Change "investigate ’unintentional transmissions’, as" to "investigate ’unintentional transmissions,’ as"
We rephrased the sentence as, “For instance, soot particles were added to investigate ‘unintentional transmission’ as they were not…”.
Page 8, Line 31: Change "was to "were"
Changed.
Page 9, Line 10: Does the author mean "tenths" or "tens?"
Thank you. Corrected.
Page 10, Line 7: Does the author mean "larger and" or "larger than?"
The latter is correct. Thank you.
Page 17, line 23: Change "grown up to" to "grown to"
Changed.
Page 17, line 24: Change "others is based" to "others based"
Changed.
Page 18, line 2: Change "the layers of the T-controlled" to "layers of T controlled"
Changed.
Page 18, line 10: Change "particles are analyzed with the single" to "particles were analyzed with a single"
Changed.
Page 18, line 11: Delete "further"
Deleted.
Page 18, line 12: Change to either "using a single particle mass spectrometer" or "using single particle mass spectrometry"
The authors agree with the reviewer. The former is reflected in text.