Answers to RC1

We thank the reviewer again for reading and commenting the manuscript in such a detail. We hope that by addressing the comments, our manuscript has improved and is now suitable for publication.

The reviewer's comments are in blue with our answers in black. Extracts from the original manuscript are presented in *italic* and changes in *italic green*.

Major Comments:

1. Thank you for clarifying the reasoning behind the 0.45 µm split for the large versus small ice residual concentrations. Since the authors do not want to separate submicron vs supermicron particles, I would suggest dividing the distribution at 0.5 µm instead of 0.45 µm. As mentioned in this manuscript, this is a common place to separate the aerosol distribution when comparing to INP concentrations, so that would make the values in this study directly comparable to many other measurements and parameterizations.

We agree that for readability and for the comparison to correlation coefficient analysis a value of 0.5 µm would be better. However,the separating size of 0.45 µm is based on the observation of a sudden change in particle concentration at this size, not at 0.5 µm, thus we would like to keep it.

Minor Comments:

1. Line 131: The Methods Sec. 3.1 says each scan is 2 hr, but it is listed as 1 hr here.

Indeed, it is two hours, thank you for this comment. We correct the statement in line 131:

„During this campaign, the temperature of PINE was continuously varied between –22 and -32 °C, resulting in temperature scans with a time resolution of approximately two hours."

2. Line 134: PINE has an aerosol D50 of 4 µm, and cloud droplets may be similar, depending on the temperature, but are expected to evaporate before entering the PCVI. But it is not mentioned anywhere what the minimum size of ice crystals can be? Can they overlap with the aerosol size distribution at all?

In PINE, it is assumed that most of ice crystals form via immersion freezing (Möhler et al., 2021). This means that initial aerodynamic diameters of freshly formed ice crystals in PINE are as large as the droplets they originated from, which are in the size range of aerosol particles. Depending on temperature, supersaturation and residence time, these ice crystals grow inside PINE before leaving the instrument and entering the PCVI body. Thus, ice crystals might be as small in size as aerosol particles (with regard to aerodynamic diameter). However, we never measured the aerodynamic size of ice crystals at the exit of PINE, and therefore would not like to make a statement in the manuscript regarding the minimum size of ice crystals.

3. Line 180-181: Thank you for clarifying how the PCVI concentration factors were calculated. Fig. A5 indicates there are transmission losses of larger particles, so the PCVI net concentration factor would not be the same across all particle sizes, correct? The Authors' Response to Minor Comment #25 states:

"We did not determine the size-dependent concentration factor specifically." This is fine, but would be good to state explicitly in the Methods.

We agree and add the following sentences to the methods section 2.4:

„The PCVI concentrates the ice particles (and thereby the residuals) by a factor of total inlet to total outlet flow. This concentration factor is, due to the nature virtual impaction (e.g., Sioutas et al., 1995), dependent on the size of the particles. However, this size-dependency was not determined specifically in this study.“

4. Lines 205-206: The fact that large aerosol particles (>4um) may be transmitted through the PCVI should be mentioned again in Methods 3.3.2, when the possibility of cloud droplets being sampled by the PCVI is considered.

The reviewer is right, as a D50 of 4 µm is not meaning that no particles larger than this size are transmitted. We add the following statement to section 3.3.2 (results and discussion):

„Please note that a certain fraction of aerosol particles larger than 4 µm can be transmitted in PINE, however, we believe that this contribution is little as the D50 of PINE is at 4 µm and the ambient concentration of such large particles at the site is quite low."

5. Line 238: Any reason or citation for the 72hr back trajectories?

This is simply a best estimate for a time period when aerosol particles from a local dust source might reach the site without experiencing major settling losses. We agree that a comparison of different back trajectories (e.g., 10 days, 5 days, 3 days) might have been useful, however, given the length of the manuscript, we did not want to include such a comparison.

6. Lines 375-377: Based on the Author's Response (Minor Comment #21) I think the final sentence of the paragraph was meant to be removed?

Thank you, yes, we remove it now!

7. Paragraph starting at line 385: Both studies mentioned looked at correlations with particles >0.5 µm, instead of >1 µm, as in this study. Consider mentioning this, as this may be driving some of the differences in correlation coefficients observed.

We agree and update the respective paragraph:

„E.g., DeMott et al. (2010) investigated the relation between INP number concentration from global measurements and particle concentration > 0.5 µm and found values between 0.6 and 0.8 for temperatures between -11.5 and -33.5 °C. At an high-altitude

station, correlation coefficients with values between 0.4 and 0.6 were found for INP concentration at -31 °C and particle concentration > 0.5 μm (Lacher et al., 2018b). One driving factor of lower observed correlation coefficients in these studies might be the smaller size (0.5 μm as compared to 1 μm), suggesting that larger aerosol particles such as dust contributes considerably to the INP population in general and especially at SPL during the respective months."

8. Lines 418-420: The expansion/flush ratio does not tell you which particles contribute the most to the INP population (concentration). Subtracting the flush concentration from the expansion concentration, integrated over each size range, would tell you which size category contributes the most to the INP concentration. Based on Fig. 10a, I suspect the smaller size category has a larger contribution to the INP concentration, as also discussed in the next paragraph. I suggest either removing the last sentence of the paragraph or changing it to something like "This indicates larger aerosol particles are more efficiently activated into ice crystals than small aerosol particles."

As the reviewer suggests, Fig. 10 panel b shows the size distribution of the residuals, meaning the flush flow concentration subtracted from the expansion flow, where it is visible that the residual concentration in the smaller size category is higher, while larger particles are mor efficiently activated into ice crystals.

For avoiding misunderstandings, we change the text to:

„The larger particles have a greater enhancement during the expansion mode, when ambient particles, cloud droplets and ice crystals can be present, relative to the size distribution in the flush mode, which is the size distribution of the ambient particles only. Thus, super-micrometer sized particles are more efficiently activated into ice crystals than smaller aerosol particles."

9. Lines 805-807: Since the Author's Response (Minor Comment #34) indicated "The supersaturation in PINE during expansion is not yet completely characterized, therefore this value (i.e. S<1.01) is only an assumption.", perhaps this discussion could include what cloud drop sizes would be expected for larger S, say S=1.05 or S=1.1, which can easily be reached in related instruments (ie CFDC). This would help to bound the range of possibilities.

At higher supersaturations (S=1.05), cloud droplets would indeed be substantially larger. E.g., for the example given in the appendix (Fig. A7), cloud droplets would reach a size of 12 μm at S=1.05, and 17 μm at S=1.1. However, given that supersaturation in PINE is established in a physically quite different way as compared to CFDCs, such high supersaturations are unlikely.

We include this now in the respective discussion in the appendix:

„Moreover, at higher supersaturations, cloud droplets could grow to larger sizes. E.g., for S = 1.05 and at -29 °C, cloud droplets could reach a size of 12 μm (17 μm for S = 1.1). However, we believe that the supersaturation in PINE once cloud droplets formed is below 1.01."

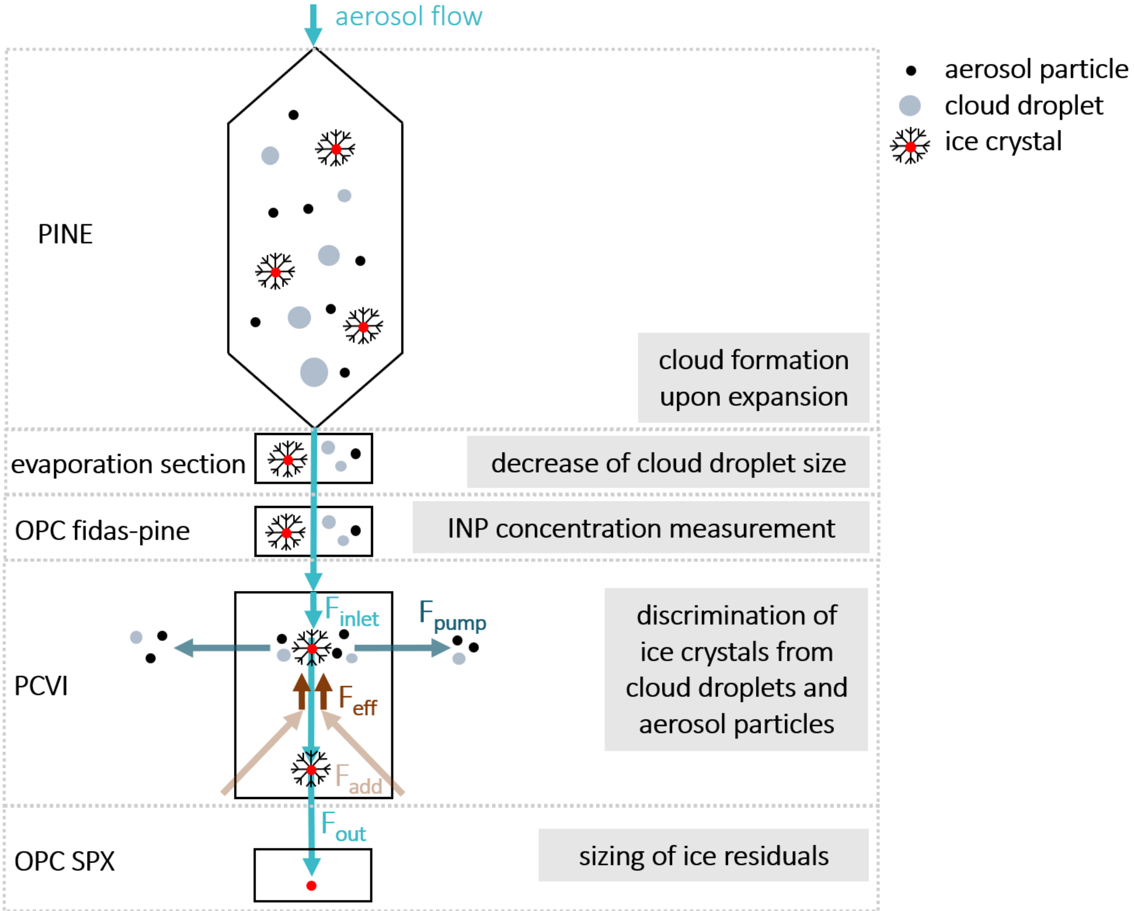
10. Some grammatical comments are given in the attached manuscript.

Thank you for carefully reading and commenting the manuscript again, we include most comments.

Figure/Table Notes:

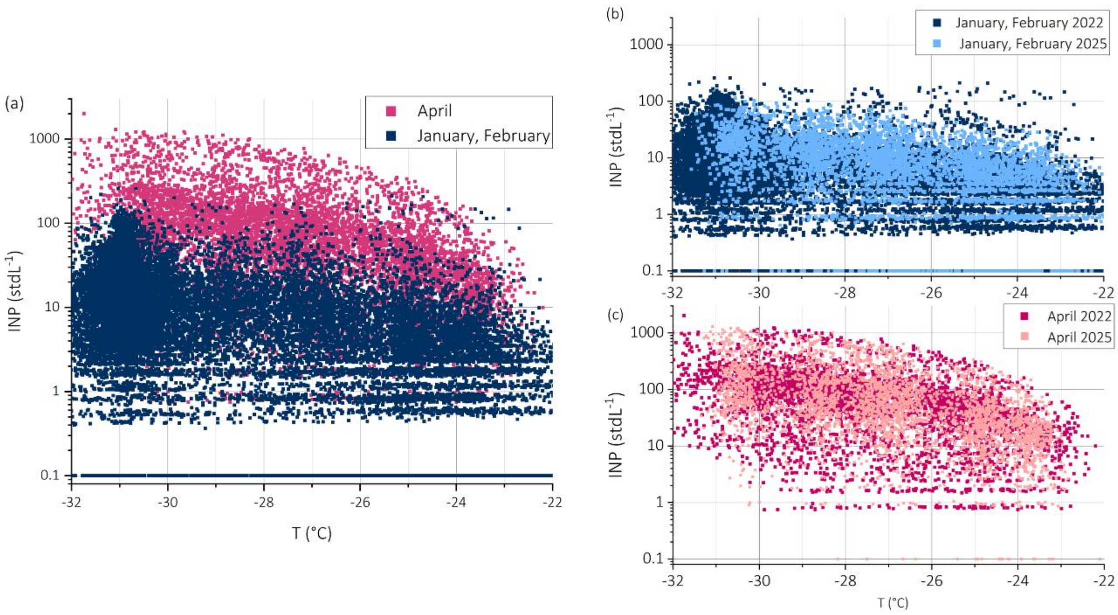
1. Figure 1: Suggest changing the text in the PCVI section to “"discrimination of ice crystals from cloud droplets and aerosol particles"

Agreed and changed.



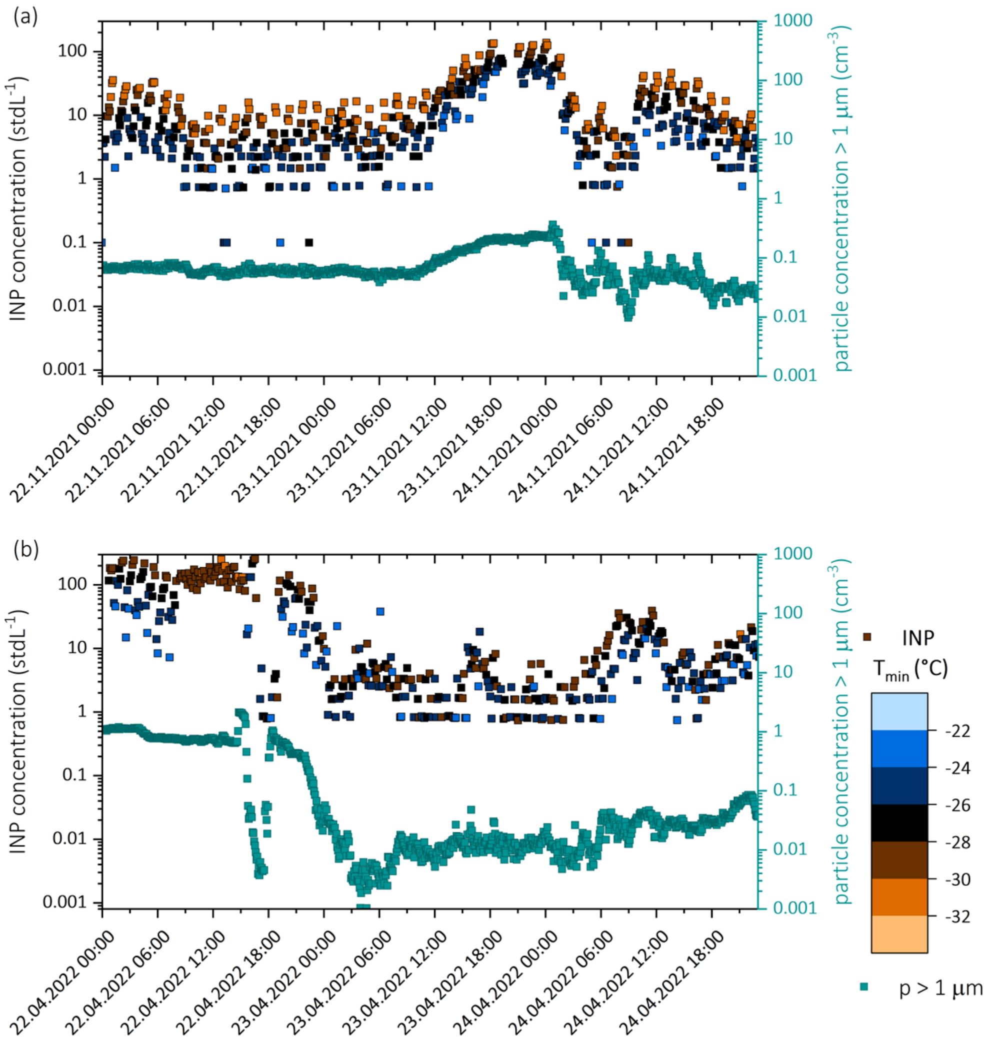
2. Figure 5: The legend symbols indicating the color of each month are very small, could they be increased in size?

Agreed and changed.



3. Figure 8: The right-hand y-axis color is very different (much lighter and more blue) than the symbols for particle concentration >1 µm. Could they be made to match?

Agreed and changed.



4. Figure 10: I still do not understand what “Sizes are reported as Polystyrene Latex particles diameter used for calibration.” means. Is the x-axis reporting optical sizes measured by the SPX for PSLs? Or have the optical diameters been converted to physical diameter (assuming spheres) using a PSL calibration based on their optical sizes in the SPX versus their physical size?

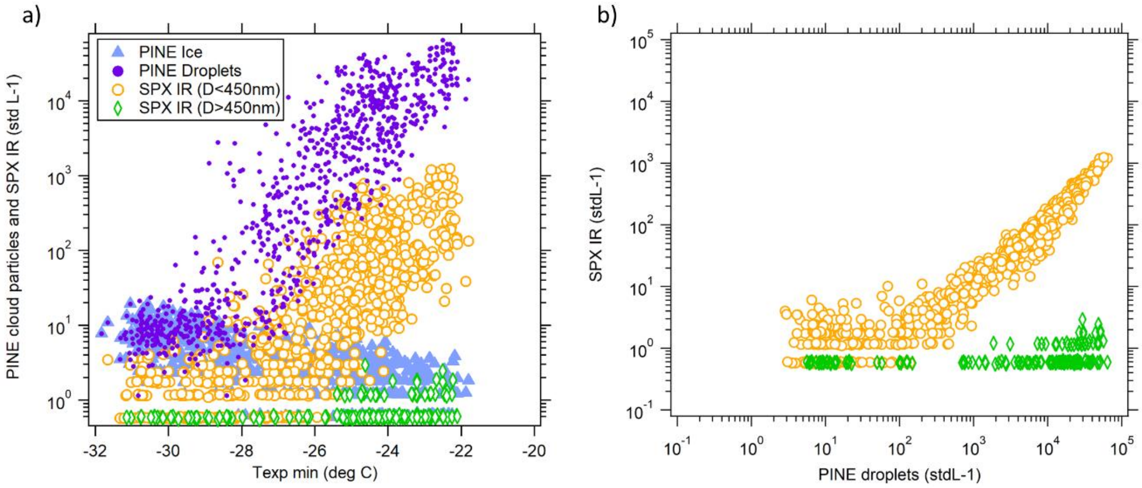
The SPX was calibrated using Polylstyrene Latex Spheres, and sizes are reported as optical diameters assuming spherical particles. We update the sentence:

„Sizes are reported as Polystyrene Latex particles diameter used for calibration and sizes are reported as optical diameters assuming spherical particles.“

5. Figure A6: I’m a little confused about what is shown in panel b. Are the blue, unfilled triangles in panel b the SPX IR (<450nm) at colder temperatures (<-28C), or are they PINE Ice crystals, but a slightly different symbol than panel a? I wonder if perhaps a legend is missing from panel b? In general, I do not see any way to determine which symbols in panel b occur at what temperature (or in which temperature range), so it is not possible to assess whether “A strong correlation is observed between small residuals and droplet

concentration (at larger than 100 droplets stdL-1), corresponding to temperatures above -28 °C”.

We included ice crystal concentrations (blue marker) in an attempt to investigate a correlation between ice crystals and cloud droplets, but agree that it is not meaningful to show them. We exclude the blue markers now in panel b, and update the statement about the temperature above -28 °C.



„Figure A6: Comparison of PINE residual concentrations (PINE Ice) with PINE cloud particle concentrations (PINE Droplets) during a series of 909 expansions on Jan 15-16, 2022 where the minimum temperature was varied from -22 to -32°C. Panel a) Both small residuals (SPX IR (D<450 nm) and PINE droplet concentrations rise with increasing temperature (panel a). Panel b) A strong correlation is observed between small residuals and droplet concentration (at larger than 100 droplets stdL-1), corresponding to temperatures above -28 °C which is visible in panel a, indicating that the PCVI is transmitting droplets as well as ice. At temperatures below -28 °C, small residual concentrations become constant and represent the combination of small INP and interstitial aerosol breakthrough in the PCVI. Residual concentrations are corrected by the PCVI concentrating factor.“

References

Sioutas, C., Koutrakis, P., Ferguson, S. T., and Burton, R. M.: Development and Evaluation of a Prototype Ambient Particle Concentrator for Inhalation Exposure Studies, Inhalation Toxicol., 7, 633-644, 10.3109/08958379509014470, 1995.