

## Answer to Reviewer 1

We thank the reviewer for his/her helpful and detailed comments and suggestions, and believe that with the corrections made our manuscript has improved significantly.

The reviewer's comments are in *blue*, and our answers in black. Sections from the original manuscript are presented in *black italic* and corrections in *red italic*.

In this paper, the authors present the ambient ice-nucleating particle (INP) number concentration obtained using online and offline measurement techniques during PICNIC campaign at Puy de Dôme. Intercomparison between online and offline instrumentation, as well as the impact of sampling site and sampling setups for offline instruments were assessed. The authors addressed the necessity of online and offline INP measurement instruments nicely. Such instrument intercomparison is essential and of great significance for the ice nucleation and atmospheric community and requires a lot of effort. Therefore, the paper fits the scope of ACP.

However, the quality of the paper should be improved before acceptance for publication in ACP. There are several typos and inconsistent usage of abbreviations in the manuscript. The reviewer tried to go from line to line to edit the manuscript, but the authors hold the responsibility for a thorough typo, format, and grammar check before re-submission.

### Major comments

1. Residence time is a critical factor that can affect online INP concentration measurement. The discussion of different residence times for different online instruments requires elaboration.

The reviewer is right that a discussion about the difference in residence time is missing in the manuscript. While the residence time of the CSU-CFDC is mentioned in section 2.2.1

*„The residence times of aerosols in the supersaturated region are 5 s for the flow rate used (1.5 LPM; liter per minute).“*

, we add the residence time in SPIN to section 2.2.2

*„Aerosols are fed into the chamber at a sampling rate of 1 LPM and constrained to a lamina center-line with 9 LPM of sheath air. **The residence time of the particles in the chamber is 10 seconds.**“*

And a discussion related to the ice crystal size in the same section:

*„Although SPIN is operated at a lower supersaturation as compared to the CSU-CFDC, the ice crystal have a longer residence time (10 seconds) such that they grow to sizes larger than 5  $\mu\text{m}$ .“*

, and included the following description about the residence time in PINE in section 2.2.3

*„ In the PINE instrument, the residence time of aerosol particles at supersaturated conditions or in supercooled droplets is more variable as compared to CFDC instruments. The time during which cloud droplets are present during an expansion is 33 seconds. However, it should be noted that this is an upper limit for the residence time, as ice crystals formed by INPs are detected during the whole expansion period and each INP has its own trajectory within the cloud chamber.“*

Moreover, we add the following discussion about the difference in residence time to section 3.1 (*Intercomparison of online instruments*):

*„Please note that the residence time of SPIN is longer (10 seconds) as compared to CSU-CFDC (5 seconds), however, we believe that other factors such as the difference in supersaturation are more important here.“*

*„In addition, the residence time of particles in PINE is not as well quantified as in the CSU-CFDC, and might be longer (maximum 33 seconds), that might impact INP concentrations.“*

2. The logical flow of the introduction section needs to be more organized.

We hope that the revised introduction is more organized.

3. The presentation quality, for example, marker colors in figures, should be reconsidered.

As the reviewer is not specifically pointing out which figure or table he/she is referring to, we would not like to make changes. With many instruments and cross-comparisons, we have done our best to clearly distinguish measurements using point types and colors.

4. The mixed-use of units and abbreviations is problematic.

By addressing your and the second's reviewer comments regarding this, and by carefully checking the manuscript, we hope that all abbreviations and units are used properly now.

#### Specific comments

L4: ice nucleating particles -> ice-nucleating particles.

Corrected.

L5: It would be more informative for the readers if the authors could provide numerical ranges of the temperature range and the orders of magnitude here.

We include the temperature range now, but the orders of magnitude were already given.

*„...the relevant temperature range for mixed-phase clouds (< -38 °C) covers up to ten orders of magnitude,..."*

L10: What are the wall temperatures of the two CFDCs in immersion freezing mode at -5 °C?

We corrected the sentence, as no online INP measurements were performed at temperatures warmer than ~ -20°C

*„INP concentrations were detected ~~online~~ in the immersion freezing mode, between ~ -5 °C and -30 °C.“*

L13: "temperatures below -20 °C" contradicts with the statement in L10.

Agreed and changed (see comment above).

L16: Missing a comma before INDA.

Corrected.

L22: It's better to clarify these are temperature spectra.

Corrected.

*„...and the obtained INP freezing spectra were compared at 1 °C steps. ...“*

L23: Not all offline instruments were operated at 1 °C step according to Table 1.

The cooling rate (temperature change per minute) is not the temperature interval at which the instruments are compared; the cooling rate describes the change in temperature during the cool-down process. Indeed, not all the instruments were operated at a cooling rate of 1 °C per minute, as described in Table 2 (formerly Table 1), however, the results of the different instruments are compared at a 1°C step.

L25-27: The explanation of the discrepancy between SPIN and CSU-CFDC is confusing. CSU-CFDC is also a CFDC-type instrument, isn't it?

Yes, both the instruments are CFDCs. We are explaining this discrepancy in more detail in the results and discussion, as we wanted to keep the abstract as short as possible.

L30-31: The description of the offline sampling technique adds little to the results. Either combine it with the description between L13-15 or remove it.

We deleted the sub-sentence in lines 30 – 31.

L33-34: Did the authors collect filter samples simultaneously on the rooftop and in the laboratory? Fig. 1 shows that IS and LINDA analyze filters from the rooftop, and the rest five offline instruments analyze filters collected in the laboratory.

Yes, those measurements were all performed simultaneously, as written in lines 21 and 22. We also add to line 34 now:

*„... compared to measurements performed **simultaneously** behind the whole air inlet system.“*

L36: Do the authors mean primary ice formation?

Yes, „*The first formation of ice...*“ refers to primary ice formation.

L53-55: Could shorter sampling time for offline measurement techniques result in a smaller sampling volume, and therefore INP concentration below the detection limit?

Yes, the same sample flow with a shorter sampling leads to a lower or even zero number of very rare INPs on the same filter; we include this statement now:

*„Results from offline INP measurements can also be obtained for shorter periods, however, this impacts the limit of detection and may lead to a lower or even zero number of very rare INPs.“*

L58: By saying organic INPs, do the authors mean biological INPs here? It would be beneficial for the readers if the authors can add a few lines here to briefly discuss the sampling size limit of most online instruments, which normally excludes pollens and dust particles above 10 - 20 µm that are ice-active at warmer temperatures and could be captured by offline sampling.

Organic refers to biological and biogenic material. We discuss the discrepancy caused by an incomplete sampling of online and offline techniques detailed in lines 73 – 103.

L70-72: Please reword.

We changed the sentence to:

*“Below -10 °C, instruments showed good agreement using SNOMAX® and natural dust samples. Discrepancies occurred using SNOMAX® above -10 °C, with illite NX above -25 °C, and with potassium feldspar between -20 and -25 °C.”*

L74-87: The logic can be improved here. The statement here is a general comparison between the size ranges of online and offline sampling techniques, instead of “the size range of aerosol particles that are INPs”. These lines should be combined with the paragraph before, please refer to the previous comment on L58. Following the impact of aerosol type and nucleation temperature in instruments’ comparability at the end of the last paragraph, a review of previous intercomparison results showing the impact of aerosol size range on INP concentration measured by different instruments is missing here, which is helpful for the readers.

INP can have different sizes and it is not trivial to infer them, especially in ambient air. As we do not infer the size of the INP in this study, we would like to only give this general statement about the sampling size range of the online and offline techniques, and the general size range of the dominant INPs.

We also reference other intercomparison studies in the laboratory and the field controlling the size range of aerosol particles now:

*„In laboratory-based intercomparison studies, it was suggested that generally good agreement between methods was achieved by controlling the aerosol particle size distributions used for the INP experiments (Wex et al., 2015; DeMott et al., 2018; Burkert-Kohn et al., 2017). At ambient conditions, however, aerosol particles and INPs can span a wide size range, which can be crucial for determining the real ambient INP concentration, and for comparing INP measurement techniques that cover different size ranges (Knopf et al., 2018).*

L88: a -> an.

Corrected.

L102-103: Is this statement relevant to instrument comparison?

In the light of the potential non-sampling of large INPs using online instruments, it is, in our opinion, an important aspect of intercomparison studies in ambient air, in close proximity or further away from emission sources of large particles.

L104-107: Consider replacing “Moreover”. Is this paragraph relevant to instrument comparison? If yes, please organize the logic.

We rephrased the paragraph to:

*“Ambient INP concentrations can be close to typical instrument detection limits (Boose et al., 2016a) and the way measurements close to detection limits are considered for averaging INP concentration over longer sampling intervals, which can be done for comparing different instruments, is another important aspect of making ambient measurements. Ambient INPs show a wide range of concentration across the relevant temperature range (e.g., Kanji et al., 2017), and it should be ensured that even low numbers of INPs, close to instruments’ detection limits, are captured.”*

L114: Move the definition of CSU-CFDC to L111 after the first appearance of “online instrument”.

Done.

L115: Please reword.

We changed the sentence to:

*„However, a high bias for offline methods, sampling particles onto filters or into a bulk liquid, against an online method was observed below -20 °C.”*

L130: What types of cloud form and occur at Puy de Dôme? Is it liquid clouds or mixed-phase clouds? If they are liquid clouds, how are the aerosols connected to mixed-phase clouds?

Both liquid and mixed-phase clouds occur at Puy de Dôme, however, the cloud phase was not determined during the PICNIC campaign. Mountainous regions typically act as a pump for air masses into more elevated levels of the atmosphere, thus our measurements of INPs at the Puy de Dôme station are seen as a potential INP population that can be lifted higher up in the atmosphere.

L136: was -> were.

Corrected.

L138-139: Consider removing the statement.

To point out differences as compared to other studies, we would like to keep this statement.

L150-151: Please specify the size range of SMPS. Why do the authors couple an SMPS to a CPC?

The size range of the SMPS is 10 to 560 nm, we include the size range now in the text. As this is a custom-made SMPS, we mention the CPC model used to measure the number concentration of the size-selected particles. We re-formulated the sentence to:

*„The submicron aerosol particle size distribution was measured using a custom-made scanning mobility particle sizer (with a particle diameter range from 10 – 560 nm) operated with a condensation particles counter (CPC, model 3010, TSI)“*

L155-156: Please elaborate on the characterization of WAI. How was the transmission efficiency computed, using number or mass concentration? What instrument was used to measure the concentration of 10 µm particles? Can this explain the consistently higher INP concentration from the filters sampled from the rooftop? Did the authors measure filters collected on the rooftop and downstream of the WAI during the same period with the same instrument and check the difference?

The transmission efficiency of the WAI is inferred by calculations, no measurements were performed for characterizing it. We include now the references for those calculations:

*„Moreover, the transmission efficiency of the WAI is dependent on wind speed. Calculations show that at values of 7 (10) m s<sup>-1</sup>, 93% (84%) of the particles with a diameter of 10 µm are entering the inlet (Hangal and Willeke, 1990; Baron and Willeke, 2002).“*

As discussed later in our text, this might explain discrepancies, in combination with a breakup of larger aerosol particles within liquid suspensions.

*„Calculations of particle transmission efficiencies reveal that the majority (>90%) of 10 µm particles are sampled at the WAI. Next to this potential non-sampling of larger aerosol particles, it is also conceivable that in-suspension fragmentation/disaggregation of especially larger particles, which were more often sampled on the rooftop, results in an elevated INP concentration, as discussed in DeMott et al. (2017)“*

We sampled filters at the rooftop and at the WAI simultaneously and analyzed them with IS (rooftop) and INSEKT (WAI). As INSEKT is a re-built of IS, we concluded that differences do not mainly come from instrumental differences.

L157: Delete “, and”.

Done.

L180: The data points are already very limited (only 20 and 34 points according to Table 2) for a 14-day measurement period with a time resolution of 10 min. How could this be?

First, this is because the data from the online INP instruments were compared at temperature intervals of ± 1 °C and overlapping sampling time. As the CFDCs are not automated and continuous operating instruments, daily sampling times were approximately two times 4 hours; each sampling period is thereby 10 minutes between two 5-minute filter background measurements. Second, for some time during the day, SPIN was operated at colder temperatures, relevant for cirrus clouds (Wolf et al., 2020). Moreover, as stated in the manuscript, due to a temperature calibration of PINE after the campaign, fewer data points were available at the temperature chosen for the intercomparison.

*„It should also be pointed that, due to a temperature calibration performed after the PICNIC campaign, the PINE had fewer overlapping measurements with CSU-CFDC as initially targeted.“*

L181-183: What is the flow rate of PFPC in this study? What type of impactor(s) was used?

The flow rates were the same as in Gute et al. (2019), as well as the impactor, which is part of the instrument. We include this now in the text:

*„The PFPC was deployed at a separate inlet and used an impactor with a 50% size cut at 2.5  $\mu\text{m}$ . The inlet and outlet flow of the PFPC were kept at the same values as described by Gute et al. (2019), i.e., 250 LPM and 10 LPM, respectively.“*

L186-192: What are the INP concentration factors for the other two online instruments? These values should be reported even though the authors decided to use 11.4 for all three instruments. Do different geometries and structures of the online instruments, as well as the impactors installed at instrument inlets affect INP concentration factors? An assessment is needed.

As SPIN is operated with a 2.5  $\mu\text{m}$  impactor, no major difference to the CSU-CFDC is expected during times sampling and not sampling at the concentrator. PINE is operated without an impactor when sampling not at the PFPC, such that variability might arise in the INP concentration factor. We include now the following statement:

*„For the intercomparison between the online INP instruments, the same INP concentration factors were applied for simultaneous measurements. This did not have an impact on the instruments' comparability, given that the instruments did not use additional impactors smaller than the PFPC's impactor with a size cut of 2.5  $\mu\text{m}$ . The INP concentration factor used for the online intercomparison is thereby a campaign average of 11.4 and has a standard deviation of 1.7. This INP concentration factor was inferred by consecutive measurements with the concentrator turned on and off sequentially, using CSU-CFDC, which performed such measurements most frequently. The average concentration factor derived with PINE was similar (campaign average 10.9) but with a higher standard deviation (5.8), that might arise from the fact that PINE does not use an impactor when not sampling at the concentrator, such that larger particles, that are ice-active, can enter the instrument and contribute to more variation of the measured INP concentrations.“*

L206-208: Please specify the size range of the OPCs for the three online instruments. What's the transmission efficiency of the single-jet impactors? Do the impactors modify the sampling flow or aerosol population entering CSU-CFDC compared to the other two online instruments?

As the size range of the OPCs is a detail that is very instrument-specific, and do not impact the interpretation of the results, we do not see the need to state the size ranges of the OPC.

The CSU-CFDC has a 90% transmission efficiency from below the OPC lower size bin to the impactor size (DeMott et al. (2017), which is comparable to PINE (90%; Möhler et al., 2021) and SPIN (100% below 2  $\mu\text{m}$ ), thus we do not think that this impacts the comparability of the instruments.

L211: What's the sample RH entering CSU-CFDC?

As described in prior publications (e.g., Rogers et al., 2001; DeMott et al., 2017), diffusion dryers are used to reduce the RH in the sample air upstream of the impactors to immeasurable values (e.g., less than 5%), which implies a dew point temperature typically below -20  $^{\circ}\text{C}$ , preventing any chance for spurious supersaturations as the air cools to the steady state RH of the CFDC.

L240: Why do the lamina supersaturation and  $\text{RH}_{\text{water}}$  have different uncertainties?

The uncertainties are the same. We have corrected this error.

*„The supersaturation employed for this study was  $2.8 \pm 2.5\%$  ( $102.8\% RH_{water} \pm 2.5\%$ )...“*

L241: What's the sample RH entering SPIN?

SPIN used diffusion dryer tubes filled with silica gel and molecular sieves upstream of the sample inlet. This brought the sample RH to below 20%.

L242: What's the transmission efficiency of the impactor? Will it change the sampling flow and aerosol population entering SPIN compared to the other two online instruments?

SPIN used a cyclone impactor with a D50 size threshold of 2.5  $\mu\text{m}$ . Below 2  $\mu\text{m}$ , the reported aerosol particle transmission efficiency is 100%. The use of the impactor should not change the sub-2  $\mu\text{m}$  aerosol population sampled by SPIN as compared with that sampled by other online instrumentation.

INP concentrations measured with SPIN are directly compared to the CSU-CFDC, that uses an impactor with the same size cut, thus, we do not expect relevant discrepancies arising from this.

PINE is operated without an impactor and has a 50% transmission efficiency of particles < 4  $\mu\text{m}$ , as stated in section 2.2.3, such that differences might arise.

We include such a discussion now at the end of section 3.1:

*„It should be noted that differences between the online instruments might arise from the difference in impactors. CSU-CFDC is operated with two single-jet 2.5  $\mu\text{m}$  impactors, while SPIN is using only one, and PINE is operated without an impactor and thus has a 50% aerodynamic size-cut at 4  $\mu\text{m}$  due to the loss of particles in its inlet.“*

L252: Application of correction factor(s) may improve the systematic underestimation of SPIN compared to CSU-CFDC. Consider moving the clarification between L525-527 here.

We clarified our reasons for not using correction factors now:

*„As the degree of aerosol lamina spreading was not quantified in this study, no correction factor was applied.“*

L255: either side-> both sides

Corrected.

L258: Please keep consistent use of StdL or L in the paper, for example, L270, L274, and L293. Please check the figures and text throughout.

The description of the PINE instruments refers to volumetric flows (Möhler et al., 2021), e.g., during the expansion the volumetric flow is kept constant, thus the usage here of L is correct. Only when we refer to standard liters, we use StdL. E.g., the results of INP concentrations are given in stdL, in order to be able to compare our results to other measurements.



To clarify that all INP concentration measurements are reported in numbers per liter of air at standard conditions (stdL), we now added the following statements at the beginning of the online and offline sections:

*„Three different online INP instruments were operated behind the WAI in parallel for several hours per day. INP concentrations were determined for single particles activating in a temperature range between ~ -20 °C and -30 °C, in the condensation/immersion freezing mode (via controlling processing relative humidity). All INP concentrations are referenced to standard liters sampled.“*

*„For offline INP analysis, aerosol particles were collected simultaneously with the different sampling setups during 8-hour intervals. ). All INP concentrations are given with reference to standard liters sampled.“*

L262: Please include “e.g.” in the citation or complete the list of citations.

There is only this study about cirrus-relevant INP concentration measurements during the PICNIC campaign using SPIN. Thus, the usage of „e.g.“ would not be correct.

L292: *welas-2500 OPC*, delete “which”.

Corrected.

L293: Specify the LOD is specific for the two consecutive experiments in this study.

*„The *welas-2500 OPC* has an optical detection volume of 10%, thus has a limit of detection of 2.5 INP per liter for two consecutive experiments.“*

L299: , however -> However. Please check the usage throughout.

Corrected.

L343-345: How long did the transport take? Why are the authors so certain such exposure to heat doesn't impact the results? What are the dominant INP source at Puy de Dôme?

Yes, the reviewer is correct, and we should be more cautious here. We rephrased this to:

*„The samples were not actively cooled during transport, however, given the relatively short travel time of ~ 8 hours to the laboratory in Frankfurt, we do not consider that this impacts the results, but cannot be excluded for certain (Beall et al., 2020).“*

No study to identify the dominant INP source at Puy de Dôme exists. A PINE instrument is installed permanently at the site now, such that in the future this question might be addressed in more detail.

L347: (2015 -> , 2015

Corrected.

L387: 100 nm deionized water?

Corrected to:

*„...two 100 nm -filtered deionized (DI) water rinses...“*

L415: Are there SEM images or other evidence to indicate “release of all particles from the filter”?

This is an interesting question and might be relevant for all filter-collection based samples. However, we did not perform such measurements for this ambient INP intercomparison study but are planning to perform these characterizing experiments for a separate study.

L426: Is “real insects” opposed to plastic or resin insects? “insects” would be sufficient.

We deleted „real“.

L429: sec. -> Sect. Please refer to ACP guidelines.

Corrected.

L443: Does the camera illuminate the droplets?

Yes, the light below the tubes illuminates the tubes containing the solution.

L450: Define “UNAM”.

We defined it now in the section title 2.3.7

*„2.3.7 **Universidad Nacional Autónoma de México** Micro-Orifice Uniform Deposit Impactor–Droplet Freezing Technique (UNAM-MOUDI-DFT).“*

and corrected the abbreviations in the text and figures.

L461: Please explicitly indicate aerodynamic particle size here.

Included now:

*„...determines the concentration of INPs as a function of temperature and **aerodynamic** particle size...“*

L465: x -> ×

Corrected.

L472: It’s very nice to indicate the units of all physical quantities here. The unit for INP concentration is missing. Is  $f_{nu}$  supposed to be dimensionless? What is the range of  $f_{ne}$  used in this study?

The reviewer is right, the correction factors are dimensionless, which we indicate now in the manuscript:

*„...  $f_{ne}$  is a correction factor, that varies between 1.2 and 4.7, and that takes into account the uncertainty associated with the number of nucleation events in each experiment“*

L480: Delete “as”. Please check the usage throughout.

We believe that the usage here is correct.

L496-498: Please reword.

We write now:

*„However, it should be noted that also the CSU-CFDC might not **measure** the **total ambient** INP concentration, due to aerosol lamina properties and size cuts, which will be discussed below in more detail **and that can lead to an underestimation of the INP concentration.**“*

L499: Fig. 3, panel a -> Fig. 3a. Please replace other appearances accordingly.

Done.

L500-502: Could this be caused by the absence of correction factor(s)? Please refer to the comment on L252.

Indeed, we think that it is related to the aerosol lamina properties, which we write two lines after L500-502.

*„A possible explanation for this systematic deviation could be related to the aerosol lamina properties.“*

L514-515: What does this statement mean?

We clarified our statement now:

*„... ultimately this may be an issue with how the central lamina is introduced to the chamber, **and how the thermal gradients and non-laminar flow at the location where the aerosols are entering the chamber impact their spreading...**“.*

L515-523: What about the residence time difference for CSU-CFDC and SPIN?

The difference in lamina RH between the CFDC instruments makes an interpretation of any potential differences in INP number concentration due to differences in residence times convoluted.

L525: “uncalibrated” doesn’t sound right.

We write now:

*„The impact of aerosol spreading was not quantified during the campaign, and data reported for the CSU-CFDC and SPIN instruments here remain **original** to account for this phenomenon“*

L528-529: Delete “(Fig. 3, panel b)”. Add “Fig. 3” in the second bracket.

Done.

L532-535: What about the residence time of aerosols in CSU-CFDC and PINE?

Also the second reviewer was pointing out that we did not discuss the residence time of the particles in PINE, thus we added to the method section about PINE:

*„In the PINE instrument, the residence time of aerosol particles at supersaturated conditions or in supercooled droplets is more variable as compared to CFDC instruments. The time during which cloud droplets are present during an expansion is 33 seconds. However, it should be noted that this is an upper limit for the residence time, as ice crystals formed by INPs are detected during the whole expansion period and each INP has its own trajectory within the cloud chamber.“*

And add the following discussion to the part of the manuscript the reviewer is referring to:

*„In addition, the residence time of particles in PINE is not as well quantified as in the CSU-CFDC, and might be longer (maximum 33 seconds), that might impact INP concentrations. .“*

L547-550: Again, is it possible to measure filters collected on the rooftop and downstream of the WAI during the same period with the same instrument and check the difference?

We did this using two identical setups: INSEKT (measuring at the WAI) is a re-built of IS (measuring at the rooftop). To point this out, we added the following sentence to the results comparing the other offline methods to INSEKT:

*„Again, the IS and LINDA, sampling filter on the rooftop, tend to measure higher INP concentrations (Fig. 8, panels e, f), and only 27% and 19% are within a factor of 2 of the INSEKT measurements, respectively. As INSEKT is a re-built of IS, a difference due to their setup is unlikely.“*

L557: Delete “impactor”, the I in MOUDI is impactor.

Done.

L582-584: Please specify “a large temperature range”. FRIDGE and IS seem to cover even wider temperature ranges in Figs. 5-7.

We included now the temperature range:

*„This method was chosen since filter collection for INSEKT was performed in the laboratory at the WAI inlet, similar to that for most of the other methods, and since it covers a large temperature range (approximately from -8 °C to -25°C) of INP measurement.“*

We believe that INSEKT is the most suitable reference method, as it is the same as the well-established IS, which was, however, sampling on the rooftop and not via the WAI. As compared to FRIDGE, INSEKT might be more reliable towards warmer temperatures due to the larger sampling volume.

L585-586: Can the authors add the comparison between two different samplers in the appendix?

The comparison is already shown in Figs. 5 – 7, where both samplers were used to collect filters in parallel, and were analyzed with INDA and LINA. We relate this statement now to these figures.

„Figure 8 includes only data for INDA and LINA obtained from the standard filter holder, as no influence from the two different samplers (standard and HERA) were observed (see Figs. 5 – 7).“

L588-L593: Tab. 2 -> Table 2. Please refer to ACP guidelines. Does droplet size (2.5 µL vs. 11 µL) play a role here?

Corrected. We believe that the difference does not arise from the different droplet sizes, and state our reasons for this in the text:

„In addition, the methods use different suspension volumes for the INP detection. However, measurements with INSEKT and FRIDGE at the Jungfraujoch show a good agreement (Lacher et al., 2021), which indicates that the larger uncertainty in the present study was not caused by the different suspension volumes, but rather arises from the larger uncertainty in the sample flow from FRIDGE.“

T594-595: Which stage(s) and size(s) did UNAM-MOUDI-DFT use to quantify INP concentration?

For this study, particles collected from stages 2 to 7 were used, which we clarify now in the method section 2.3.7:

„Aerosol particle collection was carried out by an inertial cascade impactor (MOUDI 100R, MSP) which divides the particles according to their aerodynamic diameter in each of its 8 stages (cut sizes: 0.18, 0.32, 0.56, 1.0, 1.8, 3.2, 5.6, and 10.0 µm). For this study, particles impacted on stages 2 to 7 were used.“

L596-597: Again, SEM images before and after washing would help.

The reviewer is right that this would be very helpful measurements, however, such measurements were not performed yet. Ice nucleation experiments are sometimes performed on washed glass coverslips where the observed average median freezing temperature are close to -36°C, as shown in Córdoba et al. (2021). This means that freezing temperatures observed above -35°C are not caused by the glass coverslips. We will take into account the reviewer suggestion in upcoming studies.

L598: Puy de Dome -> Puy de Dôme.

Corrected.

L602-603: miss -> lose. By saying “impaction”, do the authors mean particle impaction on the WAI surface? nanometer-sized what? It would be helpful to perform an estimation of diffusion loss.

We corrected the sentence to:

„This could be supermicron particles, that are lost by impaction in bends, or might not be sampled especially under high-wind conditions, and nanometer-sized particles that are lost by diffusion.“

L605: Again, what's the dominant source of INP at Puy de Dôme in October?

We do not know the dominant source of INP, no study on the investigation of the INP at Puy de Dôme in October was performed, as this is technically complex (e.g., using PCVIs downstream of online cloud chambers).

P616-617: Please reword.

We changed the sentence to:

*„This indicates that the discrepancy between rooftop and WAI samples does not **only** arise from a non-sampling of larger INPs, at least those remnants in liquid suspensions **after the first freezing experiment was performed.**“*

L634: Please reconsider a more informative and precise title.

As there is no title in line 634, we assume that the reviewer means the title of the next section 3.2.1 „Comparison to aerosol particle measurements“.

We change the title now to *„Investigation of INP differences using aerosol particle measurements“*

L640: What is “the presence of aerosol particles”?

Changed to:

*„In order to get a better insight into this deviation, the time series of the difference between the INP concentration measurements from the IS (rooftop) and INSEKT (laboratory) is investigated in relation to the wind velocity and the **concentration** of aerosol particles.“*

L643: Is “lognormal difference” appropriate here?

As the measured INP concentration are a strong function of temperature and vary over several order of magnitudes between the represented temperatures of -10 °C, -15 °C, and -20 °C, it was necessary to use a lognormal difference.

L645-646: Please clarify these are number concentrations.

Done.

L647: larger than 0.5 and 1 µm -> between 0.5 – 2.5 µm and 1 – 2.5 µm.

Corrected.

L649: Does the inlet refer to WAI discussed between L604-609? If yes, how come PM<sub>2.5</sub> are mostly lost when 10 µm particles have a transmission efficiency above 60%?

This is due to the fact that those measurements are derived from the CSU-CFDC, that is operated with an impactor, which we explain in section 2.1.

L666: Please reconsider a more informative title.

We changed the title to *„3.2.2 **Comparison of INP concentrations using quartz fiber and polycarbonate filters**“*

L671-673: Please reword.

We write now:

*„While both LINA and INDA can analyze particles collected with polycarbonate filters (creation of solution using the washing water), only INDA can analyze quartz fiber filter punches that are immersed in ultra-clean water.“*

L682-683: Shouldn't this conclusion be drawn by comparing the results for INDA and LINA using quartz fiber filters, and polycarbonate filters with 200 nm and 800 nm pores?

This suggested comparison was already presented, in the above:

*“HERA was equipped with polycarbonate filters (200 nm pore diameter), and the standard sampler with the quartz filter. Figure 11 shows results from sampling intervals between the 9th (daytime) to the 11th (daytime) of October.”*

An added value of the here presented intercomparison campaign is, that it is also possible to compare to other measurements (of which all offline-methods did not vary the type of filter used during the campaign). Therefore, the presented analysis is an extension of the comparison above which would include only TROPOS instrumentation.

L689: Please specify “the examined size range”.

Corrected.

*„They reported that the collection efficiency for polycarbonate filters with 800 nm pore sizes and the flow rates used here (> 11 LPM) are above 97% for all particles in the examined size range (10 – 412 nm).“*

Figure 1:

Full names of the acronyms should be given in the caption.

Corrected.

L166: within -> with

We write now:

*„...online INP measurements are compared within a time span of 10 minutes...“*

Table1:

Instrument names should be consistent throughout. Please define UNAM-MOUDI-DFT.

We use now „UNAM-MOUDI-DFT“ throughout the manuscript, and define the abbreviation.

Figure 2:

It would be helpful to indicate the factors of 2 and 5 ranges for the INP concentration of CSU-CFDC.

For clarity, we would like to keep the figure as it is.

How are the error bars for each instrument calculated? Please elaborate.

In section 2 we described the calculation of the error bars for the CSU-CFDC:

*„Statistical significance and confidence intervals for each ambient measurement are determined using the moment-based Z-statistic defined in Krishnamoorthy and Lee (2013).*

And SPIN:

*„The uncertainty in INP concentration for SPIN represents the standard deviation during a 10-minute sampling period.“*

And PINE:

*„The uncertainty for the INP concentration is 20%, which is an upper estimate from the uncertainties of the determination of the optical detection volume.“*

The INP concentrations exceed 20 #/L at -30 °C. Do the authors have an explanation for such a high INP concentration?

This is an interesting question. One possibility is that, in general, the Puy de Dôme experiences elevated INP concentrations as it is impacted by boundary layer air. Recently, a PINE was installed for permanent operation at the site, such that a better insight into the variability in INP concentration will be investigated.

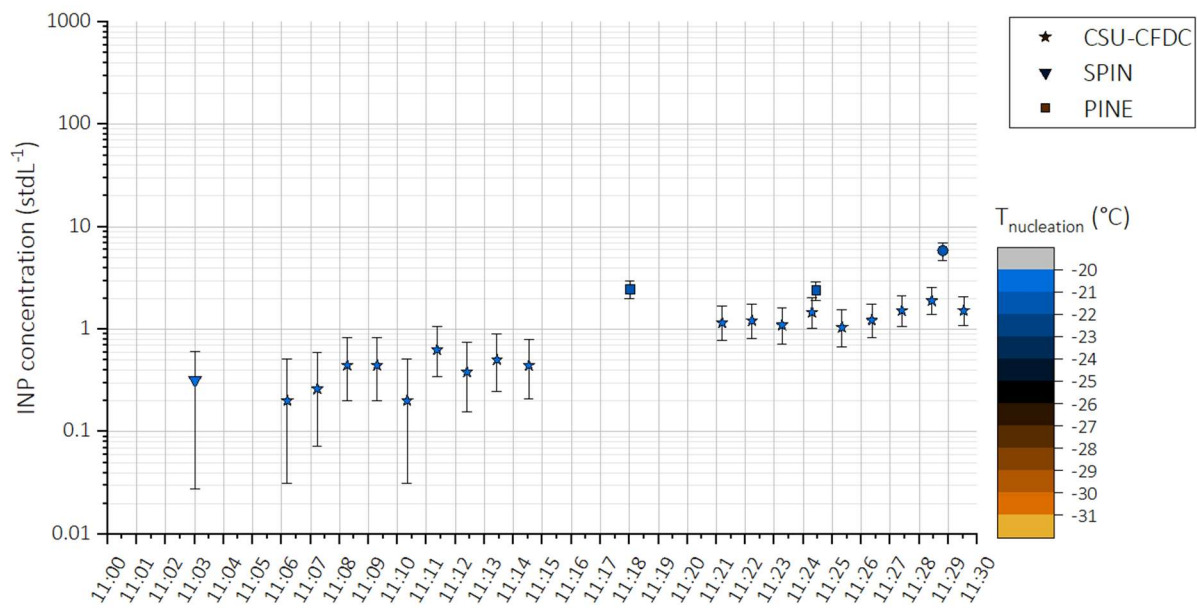
L490: Please keep the same order of instruments in the caption and legend.

Corrected.

L491: The time resolution of CSU-CFDC doesn't seem like 1 minute.

The time resolution of processed data is 1 minute, see the figure below where we show the same plot for 11:00 – 11:30:





Can the authors provide the full-time series in the appendix?

The INP concentration measurements from the full campaign will be presented in a separate publication (Freney et al., in preparation), thus we would not like to show it in this study.

Figure 3 and Table 2:

Why do the three online instruments have so few inter-comparable data points with a much higher time resolution?

Please see our answer to your question above.

Is there a specific reason for the authors to choose CSU-CFDC and INSEKT as the baselines/references for online and offline INP measurement techniques? Do the intermediate measured values by these two instruments play a role in reference instrumentation selection? L493-496 and L582-584 partly address the concern.

We specify our reasons for choosing CSU-CFDC and INSEKT in sections 3.1 and 3.2, respectively:

*„...the results from all intercomparison experiments are investigated using the CSU-CFDC as a reference instrument, given its long history of operation and good characterization.“*

*„This method was chosen since filter collection for INSEKT was performed in the laboratory at the WAI inlet, similar to that for most of the other methods, and since it covers a large temperature range (approximately from -8 °C to -25°C) of INP measurement.“*

The information in Table 2 can be merged into Fig. 3 and Fig. 8.

For a better overview of the results, we would like to keep the table as it is.

Figure 4:

Inconsistent usage of instrument name in the caption and legend.

Corrected.

The information has been included in Figs. 5 - 7?

We do not understand the question about which information should be included in Figs. 5 - 7.

Figures 5 - 7:

The marker colors for online instruments are hard to distinguish. Please consider changing marker shapes for different online instruments.

For a better distinction from the offline instruments, we would like to keep one symbol color for the online instruments.

Is it better to keep one representative panel, and move the other to the appendix?

For visualizing the agreement and the discrepancies between the methods, that vary with sampling periods, we would like to present all the panels. By showing only one panel, it might give a biased view on the results. We acknowledge that Figure 4 already shows the timeseries of the INP concentration at key temperatures, however, we would like to show all measurements at all temperatures, and in addition, the INP community is most familiar with the interpretation of freezing spectra.

Please include filter information.

We include now the information in the figure captions.

L577: missing "of".

Corrected.

Figure 8:

Inconsistent usage of instrument name in the caption and y-label.

Corrected.

Figure 9:

The symbols are hard to read.

Due to space limitations, we would like to keep the symbol size.

Figure 10:

Please change the color and label color of the right y-axis in panel d.

We do not see the need to change the label color of the right y-axis; due to the markers and the y-axis label, it is clearly defined which y-label refers to the markers.

L662: Please reword the caption.

We do not see the need to change the caption.

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