Answer to Reviewer 2

We thank the referee for commenting our manuscript and giving very valuable suggestions.

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*.

Lacher et al, do an extensive intercomparison of INP measurement techniques at the Puy de Dome research station. They generally find that all of the measurement techniques agree reasonably well, especially when accounting for instrumental differences and the sampling location (in front/behind the inlet). The manuscript is extremely well written and shows that the INP community has developed a suite of different INP measurement techniques that are capable of robustly measuring INPs in the field. I recommend the manuscript is published once the comments below are addressed.

General comments:

This is a very technical paper, which makes huge efforts to do a much needed instrument comparison. However, due to its technical nature and since it is primarily an instrument intercomparison, I wonder if it is more appropriate for it to be published in AMT. I know this is up to the authors and editor, but it is something I would consider since it mainly discussing and comparing instrument sampling discrepancies, rather than investigating processes/ atmospheric concentration of INPs.

The reviewer raises a good point, and we believe that the manuscript is suitable to be published in both journals, ACP and AMT. The reason why we submitted it to ACP is that the discussion about the comparablity is specifically taking into account that we sample ambient air, and interpret the sampling conditions such as available aerosol particle properties and meteorology that might impact the intercomparison. Please note that other intercomparison studies (e.g., DeMott et al., 2017; Burkert-Kohn et al., 2017) are also published in ACP. Moreover, we plan to submit a companion manuscript about the interpretation of the INP concentration to ACP as well. Thus, we would like to submit our manuscript to ACP, but leave the decision to the editor.

As I was reading, I often found myself wondering why the instrument intercomparison did not do more to ensure that everything was the same across measurement platforms (e.g. same filter water distributed across DFTs, same set points on the CFDCs). Then I realized that one of the strengths of the studies is that the comparison was conducted using the native sampling format of each measurement technique. I think this point could be strengthened/empahsized, as it is a very nice conclusion showing that all of the tested techniques give representative INP concentrations and are equally useable in the field.

Thank you for pointing this out. We now add the following sentences to the end of the introduction section to highlight this important point:

„*During PICNIC, seven offline techniques and three online instruments were compared over 14 days in October 2018. The aim here was to test the measurement techniques against each other in their original operation configuration, as each of them are well-established methods and were already used in several campaigns, and we wanted to create a link between these activities without changing measurement protocols.*"

At the beginning of section 4 Summary and conclusion:

„During the PICNIC campaign in October 2018, a suite of online and offline INP measurement techniques were operated simultaneously to compare the temperature-dependent INP concentrations relevant to the formation of mixed-phase clouds. The methods were deployed in their typical operation configuration without equalizing measurement setups.“

And at the end of section 4

„With regard to required precision of INP measurements to be included in models, the results from our study greatly demonstrate that the methods as used in their original configuration agreed overall well within a factor of 5.“

It is not immediately clear if all the INP concentrations are reported in $stdL^{-1}$ or not (although the figure units are stdL-1). Perhaps make it clear how this is reported especially when for some of the filter techniques (ie IS, INSEKT) a mass flow is explicitly mentioned.

We added now at the beginning of the online and offline sections, as a general remark, that all INP concentration measurements are given in standard liter.

„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.“

At a more critical level, sometimes the main messages of the paper were not very clear. First, there is a lot of discussion about why the instruments may not agree completely. I appreciate the authors taking the time to discuss these uncertainties/factors. However, all of the discussion is quite speculative and some calculations could determine if the proposed factors are the culprit or not. Second, is such a long discussion warranted when the measurement techniques agree so well i.e. majority agreement within a factor 5 or better. Along the same lines, at what point is the agreement high enough that we no longer need to do intercomparisons? Have we reached this agreement level? How much of an influence would a factor of 5 agreement have on the development of parametrizations and modelling results? Lastly, if we don't expect to get closer than a factor of 5, do we really need more field intercomparisons? Wouldn't it make more sense to do careful lab intercomparisons where we can adjust our techniques to have better agreement in a controlled setting?

Thank you for this remark. We hope that with addressing the comments from you and the other reviewer that generally the quality of the manuscript improved, and that its main message is now more clear.

With regard to the in-detailed discussion of potential discrepancies, the aim here was not only to present our measurement results but also to point out certain factors that can be a source of disagreement. However, as the reviewer points out, the discussion is speculative, as with the experimental setup used, we cannot pinpoint reasons for disagreement. Along this line, although

an agreement within a factor 5 might good enough for model representation of INP concentrations, it still is a relatively large number. Besides, we argue that the sensitivity of models to the INP number concentration is still not fully understood, and this might change when modeling different cloud types in different environments. Moreover, it is vital to investigate the reasons for this deviation, as they might have a greater impact sampling a different aerosol and INP population. E.g., the observed discrepancy between the samples collected on rooftop and behind the WAI might be greater when, e.g., sampling a dust-dominated INP population where more large particles are present that might lead to particle break-up in solution. Thus, as we do not know the factors yet that lead to discrepancies, and as setups and operation procedures of individual methods might be changed for improvement, frequent intercomparisons of methods are recommended.

While laboratory intercomparison studies are very useful to limit factors that can lead to disagreement between methods, intercomparisons of instruments that are operated in field campaigns are required, as some factors cannot be easily simulated in the laboratory, as e.g., the impact of windspeed on the transmission efficiency of particles, or simply the usage of a whole air inlet. In the lab, agreement often depends on used aerosol samples, while in the field a mixture of natural aerosol can lead to different results. Again, as we do not fully understand why there are discrepancies between instruments, we need to compare with well-known (laboratory) and ambient (field) aerosols. As most of the instruments are used for field applications, it is certainly needed to intercompare under realistic sampling conditions.

We are not aware of calculations besides the particle transmission efficiency for the WAI that could be used for determining factors leading to disagreement between the instruments.

Minor comments:

Abstract: Consider adding a discussion about the importance of INPs as this is geared for ACP, if AMT is the end journal then it is fine as is.

This is a very good suggestion. We changed the start of the abstract to:

„The formation of ice crystals in clouds is initiated by specific aerosol particles, termed ice-nucleating particles (INPs). Only a tiny fraction of all aerosol particles are INPs and their concentration over the relevant temperature range for mixed-phase clouds (< -38 °C) covers up to ten orders of magnitude, providing a challenge for contemporary INP measurement techniques.“

Line 155-156: Is there a reference for these reported transmission efficiencies?

Yes, we used calculations to report these numbers, which we clarify now and include the respective references:

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

Line 181-193: This is a bit unclear. So the concentrator was only used occasionally but how does this work when comparing between PINE, which fills for a fixed interval and then expands while the CFDCs measure continuously? Was the CFDC data excluded when PINE was filling as there would

be a different flow ratio in the concentrator when all three instruments sampled? Do you expect this to be a potential problem/ lead to a change in the concentration factors ie change in particle size distribution?

The PINE inlet flow rate is constant, during the expansion the inlet flow is maintained by a bypass flow. Thus, the flow ratios of the PFPC was kept constant. We add to the PINE method's section now:

„The flush mode is followed by the expansion mode when a valve upstream of the chamber is closed while the volumetric flow out of the chamber is set to a constant value of 3 LPM. Please note that the inlet flow rate during the expansion is maintained by a bypass flow which is the same as the flush flow rate, such that no change in the sampling flow at the WAI occurs."

Role of impactors: Does it matter that SPIN uses one impactor and CSU-CFDC is using two? The impactor cut size is distribution based so would this lead to more lager particles making it into SPIN than the CSU-CFDC? Of course this does not seem to be the case based on the results but it could be something to mention if this another source of uncertainty. I see this is now discussed a bit in the results. Perhaps it would be nice to have a table with the online instruments and their associated set up e.g., impactors, set conditions etc.

We agree that a small difference due to the use of two impactors might occur, something the first reviewer pointed out as well. Hovewer, this would lead to more larger particles entering SPIN and thus likely to a higher INP concentration as compared to CSU-CFDC, which is not observed. For clarification, we add now to the end of section 3.1 (intercomparison of online instruments) the following sentences to address the difference in the impactors used:

„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 µm impactors, while SPIN is using only one, and PINE is operated without an impactor and thus has a 50% aerodynamic size-cut at 4 µm due to the loss of particles in its inlet."

More, we appreciate the idea of adding a setup table similar to the table provided for the offline methods, and included the following table in section 2.2

Table 1: Specifications of the online instruments.

Name	**CSU-CFDC**	**SPIN**	**PINE**
inlet	WAI / PFPC*	WAI / PFPC*	WAI / PFPC*
impactor	two impactors with 2.5 µm size-cut	one impactor with 2.5 µm size-cut	no impactor; size-cut 4 µm (Möhler et al., 2021)
temperature and RH_{water} uncertainty	± 0.5°C and 2.4%	± 0.5°C and 2.5%	± 1 °C
residence time	5 s	10 s	< 33 s
supersaturation	106.5% RH_{water}	102.8% RH_{water}	> 100% RH_{water}
ice threshold	4 µm	5 µm	automated

* online instrument sampled always at the same inlet

Section 2.2.2- Is it concerning that the RHw in spin was ~3 % lower than in the CSU-CFDC yet the ice crystal detection was one micron higher? It might be worth mentioning that the longer growth

(residence) time in SPIN would ensure that at this supersaturation, the ice crystals would reach a 5 micron size even with the lower RHw than in the CSU-CFDC if this is the case. This is also discussed later but not actually calculated. Consider doing this calculation.

Indeed, due to the longer residence time in SPIN, ice crystals are growing to larger sizes as the CSU-CFDC even though SPIN is operated at a lower superaturation. To make this point clear, we add to section 2.2.2 now:

„Due to the sigmoidal shape of the impactor's size cut, OPC counts larger than 5 µm in diameter were considered as activated INPs. 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 µm."

Line 258-259: Why does the limit of detection double behind the concentrator? Aren't the background counts from the wall the same regardless of the concentrator? I this just due to having more "air" going into the instrument? This is not immediately clear, consider explaining this a bit better.

Likely our statement made was not clear here. Indeed, the limit of the detection of SPIN behind the concentrator is lower as compared to sampling not at the concentrator, as more sample is analyzed while the background counts are the same. We correct the statement such that this is clear now.

„The limit of detection of SPIN sampling at the concentration is lower (~ 0.6 INP L^{-1}) as compared to not sampling at the concentrator (~ 6 INP L^{-1}), as more sampled air is analyzed, while the ice background counts remain the same."

Section 2.2.3 -

Firstly, does this mean that PINE can only measure between -19 and -13 based on the start temperature of -13 and only a 6 degree cooling? This is not immediately clear. It might be nice to add a table with the temperatures that the comparisons were conducted at (see previous comment about impactors).

No, this is not the case, we operate PINE at temperatures *below* the frost point temperature of ~ -13 °C. Indeed, we operate PINE typically at temperatures colder than the frost point temperature. That means that the sampled air is too humid, thus we have an ice layer on the inner chamber walls, that is, however, not impacting our sampling conditions in terms of background from internally formed ice crystals. As the total water content at temperatures of below -13 °C is smaller as compared to warmer temperatures, no de-icing of the chamber walls within a shorter sampling period of a few days is needed (for a long-term operation with PINE, the chamber is de-iced frequently).

The INP concentrations were compared between -20 °C and -30 °C, as written in section 2.2

„INP concentrations were determined for single particles activating in a temperature range between ~ -20 °C and 30 °C…"

and indicated in figures 2 and 3. Thus we believe that this is redundant information for the new table 1, that we wanted to keep similar to table 2 (offline methods) and does not contain this information likewise.

Second, during the continuous expansion, aerosol particles are removed, therefore is there some sort of correction for the decrease in effective volume in the chamber? I ask as depending at the ambient pressure (I guess around 850 hPa) a 300 hPa decrease is ~35 % loss of aerosol during the expansion. This would suggest that depending on when in the cycle the set temperature is reached, a fraction of the aerosol are already removed (albeit still less than the factor of 2 threshold). Please add information about this correction if it is performed or necessary.

This is an interesting question. The intention in PINE is to analyze all aerosol particles within the volume of the chamber, which is achieved by guiding the expansion flow containing the aerosol particles and formed ice crystals over the OPC attached between the chamber and the pump. Thus, all aerosol particles that are „lossed" during the pressure decrease are measured. Indeed, the temperature that is assigned as the nucleation temperature is the minimum temperature reached at the end of the expansion (Möhler et al., 2021), meaning that a fraction of the particles were not exposed to this minimum temperature. However, we do not apply a correction factor for this, but might consider this for future studies.

Third, does the lack of impactor here also mean larger particles are expected to enter PINE than the other two online measurements since a cut size is distribution based?

Yes, this is correct, see our answers to your comment above.

Table 1: Consider adding the total volume of the air sampled for each filter technique as well as the minimum INP detection limit for each instrumental technique in the table. I know that this is corrected for, but I still think it is nice to see since the range in INP concentration covered appears to vary quite a bit.

Thank you for this suggestion. We decided to include now the limit of detection to table 2 (formerly table 1).

	Name	FRIDGE	INSEKT	INDA*	IS	LINA*	LINDA	UNAM-MOUDI-DFT
filter collection	location	WAI	WAI	WAI	rooftop	see INDA	rooftop	WAI
	time interval	8 hours (night), 4 hours (day)	8 hours	8 hours	8 hours	*same as INDA*	8 hours	8 hours
	substrate	47 mm PTFE fluoropore membrane filter, 220 nm pore size	47 mm polycarbonate filters, 200 nm pore size	47 mm polycarbonate filters, 200 nm and 800 nm pore size; 47 mm quartz fiber filters	47 mm polycarbonate filters, 200 nm pore size	*same as INDA*	15 cm quarz fiber filters	hydrophobic glass coverslips
	filter holder	custom-built semi-automated multi-filter sampling device	standard	standard, HERA	open-faced sterile Nalgene sampling heads	*same as INDA*	high volume sampler	MOUDI cascade impactor
	flow	4.8 LPM	11 LPM	standard: 12 - 37 LPM HERA: 15-41 LPM	13.5 LPM	*same as INDA*	500 LPM	30 LPM
	limit of detection (L^{-1})	4.3E-04 (8 hours)	1.90E-04	standard: 1.7E-04 - 5.6E-05 HERA: 1.4E-04 - 5.1E-05	1.5E-04	*same as INDA*	4.2E-06	6.9E-05
	filter storage	partly unfrozen	frozen	frozen	frozen	*same as INDA*	frozen	refridgerated
analysis	liquid volumes	2.5 µl droplets	50 µl suspension	50 µl suspension	50 µl suspension	1 µl droplets	200 µl suspension	100 µm droplets
	cooling rate	1 °C min^{-1}	0.3 °C min^{-1}	1 °C min^{-1}	0.3 °C min^{-1}	1 °C min^{-1}	0.3 °C min^{-1}	10°C min^{-1}

* INDA and LINA use the same collected filter

Line 343-344: Consider adding a ref where cold transportation is discussed e.g. Beall et al., (2020)

We include now the following reference:

„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 exclude it for certain (Beall et al., 2020).“

Line 369: You could add Sarah Grawe's new paper about HERA if you wanted (Grawe et al., 2023)

This is a good point, we include Grawe et al. (2023) now.

Section 3.2-

Here it would be worth mentioning the probability of detecting INPs at the warmer temperatures due to the limited sampling volumes used. It looks like LINDA and IS, which are also two of the methods using the equivalent of the most volume of air sampled, are the highest.

Agreed. We add to the beginning of section 3.2 now:

„Due to the difference in sampled volume and thus detection limit (see table 2), the probability to detect very rare INPs at temperatures above ~ -10°C varies amongst instruments.“

Line 456-458: Again it would be worth citing the importance of transporting samples frozen or at least the impact of warmer temperature transport (e.g. Beall et al., 2020).

We add now to the description of the transportation of samples for analysis with UNAM-MOUDI-DFT:

„We note that storing samples for a longer transportation time might impact the INP concentration (e.g., Beall et al., 2020). Although we did our best to keep the samples below 0°C by transporting them in a freezer with ice packs, it is very likely that the samples may have experienced temperatures slightly above 0°C right before reaching their final destination.“

Line 466: What are the impacts of such a high cooling rate? Previous studies have looked into the impact of cooling rate (e.g. Budke and Koop, 2015) and have seen a difference due to the stochastic nature of ice nucleation. This should also be discussed more in the results and not just as a conclusion. See point again below.

We agree that such a conclusion should be discussed in the results, and add now to the results section about UNAM-MOUDI-DFT:

„As shown in Fig. 8b, the UNAM-MOUDI-DFT tends to measure higher INP concentrations compared to INSEKT. This bias may be coming from the method used to capture the particles. While for the INSEKT samples Nuclepore filters were used, in the MOUDI-DFT particles were impacted on glass coverslips. A possible explanation is that not all particles are released from the Nuclepore filters. If so, this may relate to the aerosols sampled at Puy de Dome, as this bias was not seen in some prior comparisons (e.g., Mason et al., 2015). Moreover, the UNAM-MOUDI-DFT is the method using the fastest cooling rates of 10 °C per minute, such that an effect of a time dependency of ice nucleation might have impacted the results (e.g., Hoose and Möhler, 2012; Budke and Koop, 2015). However, this would have lead to an underestimation of INP concentration, such that we conclude that the ambient INP concentration are not considerably controlled by stochastic variation, or that other instrumental properties of sample colletion and analysis with UNAM-MOUDI-DFT are dominant.“

Line 495: Even though it is well known that the CSU-CFDC is a well-known and established measurement system, please add some references that attest to its established reputation.

We add now a selection of campaigns including measurements with CSU-CFDC:

„To identify potential systematic deviations between the three instruments, the results from all intercomparison experiments are investigated using CSU-CFDC as a reference instrument, given its long history of operation and good characterization. CSU-CFDC has been used extensively in laboratory intercomparisons (e.g., DeMott et al., 2011; Hiranuma et al., 2015; DeMott et al., 2018) and in a large number of field measurement studies in surface- and aircraft-based campaigns (e.g., within the last five years, McCluskey et al., 2018; Cornwell et al., 2019; Hiranuma et al., 2019, Kanji et al., 2019; Levin et al., 2019; Schill et al., 2020; Barry et al., 2021; Knopf et al., 2021; Twohy et al., 2021) over a period of more than 25 years."

Editorial comments:

Line 10: please switch to listing coldest temperature first followed by the warmer temperature when giving a T range. This should be consistent throughout the entire manuscript.

Throughout the manuscript we are listing warmer temperatures first, thus this is consistent and we would like to keep this ordering.

Line 38: Although this is a complete list, you could add studies that show a relationship between INP concentrations and cloud phase e.g. (Creamean et al., 2022; Carlsen and David, 2022; Sze et al., 2023)

Thank you for this suggestion, we added the proposed publications to the manuscript.

„The presence of INPs is important for the formation and further development of clouds since they can determine cloud phase (e.g., by a rapid cloud glaciation and associated dissipation effect; Campbell and Shiobara, 2008; Murray et al., 2012; Paukert and Hoose, 2014; Kalesse et al., 2016; Desai et al., 2019; Murray and Liu, 2022; Carlsen and David, 2022; Creamean et al., 2022; Sze et al., 2023)…"

Line 74-78: It would be nice to add a reference for this.

We added Rogers et al. (2001) now.

„Typically, online instruments, such as continuous flow diffusion chambers (CFDCs), limit the aerosol sampling to size to diameters below ~ 3 µm (e.g., Rogers et al., 2001),…"

Line 149:program-> programs.

Corrected.

Figure 1: Please have T range go from cold to warm.

See our answer above.

Line 308: remove the additional "periods".

Done.

Eq. 1: is it appropriate to call it a V_{sol} as you later refer to it as the droplets containing the suspension? Consider calling it V_{sus} or similar for suspension, but this is just a semantics thing.

We agree with the reviewer and call it now V_{sus}:

„The calculation thereby considers the volume of water used to extract the sample (suspension; V_{sus})…"

Line 347: add) at end of Hiranuma ref.

We corrected it to (Hiranuma et al., 2015).

Line 347: Consider adding that the filter had been exposed or was a sample containing filter or something similar.

We add now:

"Before starting a measurement, a filter containing the sampled aerosol was placed in a sterile Eppendorf tube, which was filled with 5 mL of ultrapure water (Rotipuran ultra, Carl Roth)."

Line 495-496: It's a bit unclear here what is meant by the true INP concentration? Does this lead to an undercounting or overcounting? My guess would be undercounting, I would specify this.

We corrected the sentence to:

„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."

Line 496: comma before which.

Corrected.

Line 500: It might be worth mentioning that an undercounting of SPIN might be expected as observed in Garimella et al, (2017). I see this is done later (see comment below), but it is interesting that SPIN would have more lamina spreading for lower RH settings than the CSU-CFDC. This could be mentioned and is consistent with Garimella et al, (2017) who reported that spreading can contribute to a factor of 10 undercounting, which is more than the factor of 3 reported for the CSU-CFDC.

We re-organized the explanation of the different factors that can impact the determination of the INP concentration in SPIN:

„Previous studies have found that the aerosol particles in at least some CFDCs are likely spreading beyond the lamina, such that not 100% of particles are in the lamina where they are exposed to the targeted supersaturation condition (DeMott et al., 2015; Garimella et al., 2017; Wolf et al., 2019). The issue of lamina spreading is likely variable and depends on the CFDC geometry, the flow conditions, and the temperature gradients between the walls, which is creating the supersaturation;

ultimately this may be an issue with how the central lamina is introduced to the chamber, and the thermal gradients existing there. Aerosol spreading causes aerosol particles to experience lower supersaturations than the target supersaturation, resulting in either a non-activation into cloud droplets and ice crystals (immersion freezing mode), or an activation into ice crystals that are not growing to sizes within the residence time in the chamber to be detected by the OPC (above the ice threshold). SPIN was operated at a lower supersaturation (2.8 ± 1.9%) as compared to CSU-CFDC (6.5 ± 1.4%). Thus, it is expected that SPIN underestimates INP concentration, by up to a factor of 10 (Garimella et al,, 2017; Wolf et al., 2020).“

Moreover, SPIN also used a larger ice threshold in the OPC of 5 µm, against 4 µm from CSU-CFDC, which has been found to impact INP concentration measurements (Jones et al., 2011). Thus, it is possible that due to a larger ice threshold size, fewer particles in SPIN were encapsulated in the intended conditions, and were less likely to reach the critical size threshold.“

Line 514: comma before which.

Corrected.

Line 520-527: Here the discussion of spreading is well discussed. I would consider reordering and only mention these issues once, like done in this section.

See our answer above.

Line 745-747: Please make sure this is also discussed in the main text as it is an interesting point to raise. Does it make sense to consider longer time scales for the importance of time-dependence at these warm temperatures i.e. maybe you need longer time scales like 5-10s of minutes for the stochasticity to matter e.g. Budke and Koop, (2015) who needed a very low freezing rate to observe a big difference (.1 K/min)?

We added a sentence in the main text, see our answer to your question before.

The reviewer is right that longer time scales might be needed. However, the intention of this study was not to investigate a potential time dependency of ice nucleation, but to compare methods used for ambient measurements, thus we would not like to add a statement or suggestion here for future experiments on a potential time-dependence.

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