Answer to the editor's comments

We thank the editor and the editorial team for their careful thoughts about the missing criteria of the manuscript to be published in ACP. We have now added discussions and statements about the atmospheric implication of our intercomparison campaign. The editor's comments are in blue, and our answers are in black. Sections from the original manuscript are presented are in *black italic* and corrections in *red italic*.

Authors,

Thank you for your revised manuscript and for your very detailed responses to the two sets of referee comments. I also apologize for the extra delay in my response. I needed to discuss the decision with the editorial leadership to make a final decision about the placement of the manuscript. The manuscript is very nice and provides an important and very useful ambient intercomparison of various INP sampling and measurement techniques, even if it is very technical. You have done a good job revising the manuscript based on the feedback, and it is generally close to being ready for acceptance. The point referee #2 introduced about whether ACP or AMT would be a better fit is an important one, and what I needed to discuss. Author guidelines for ACP research articles are now nicely summarized here (with more detail at the web address/link below):

"Implications: Discuss what the results mean for our understanding of the state and/or behaviour of the atmosphere and climate, which is the main requirement for publication in ACP. The editor's acceptance/rejection decision will be strongly guided by this component of the concluding section."
https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html

While the manuscript itself is essentially ready, the editorial team and I don't think the text yet adequately fits the requirements for publication in ACP. In particular, the abstract and conclusions need edits to make them closer to these requirements. Had I thought through these points before review, I would have suggested moving the manuscript to AMT at that point. That would require a new round of review, however, and I don't want to require that of your manuscript at this stage. So I suggest that we proceed toward publication in ACP, but following an additional round of edits to the abstract and conclusions. This final round of revision can proceed through what hopefully should be a very quick round of review only by me, and it will not require a second review by anonymous peers.

Please see the new requirements for publication, linked above. In particular, your abstract is currently at ~475 words, which is well above the new maximum of 250 words. The guide provides a very useful template of components to include, with point 7 (the importance and implications of the results) as a critically missing component for the present manuscript version. I suggest that you add some discussion of the broader atmospheric implications of the study within both the abstract and conclusions, but hold to abstract word limits, as now posted.

More specifically, the last section of your manuscript is still largely a summary of the differences between the different techniques and the technical basis and implications, which is not yet sufficient for an ACP research article. This is where the article "must convincingly demonstrate important implications for our understanding of the state and behaviour of the atmosphere and climate" and not just the implications for how measurements can best be performed or interpreted. The implications of the study are currently expressed primarily in the single sentence added during revision (Lines 836-837: "With the regard to ...").

While you will not be required to address all of these suggestions, it may be useful to address some of these points. For example:

- What does the stated factor of 5 mean for our understanding of the atmosphere?

• Can you be more detailed and specific in your discussion about how this factor of 5 would impact atmospheric models?
• Clearly state what the novelty of the current study is. How do your conclusions add to or alter conclusions from previous reports and the broader, existing understanding of atmospheric INP and/or atmospheric processes?
• The final two sentences of the conclusions ("Ambient INP intercomparison campaigns …") does not provide a powerful ending conclusion, especially given that these summary statements have been broadly known. For example, you might consider specific ways in which you would expect your results to constrain models or improve atmospheric understanding with respect to the needs and questions presented e.g. in publications like these:
o Burrows et al., 2022: https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2021RG000745
o Coluzza et al., 2017: https://www.mdpi.com/2073-4433/8/8/138
o Zamin et al., 2017: https://journals.ametsoc.org/view/journals/amsm/58/1/amsmonographs-d-16-0006.1.xml
o Ervens et al., 2011: https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2011JD015729

I am happy to answer further questions via email, if you like. Otherwise, I look forward to looking at your revised manuscript.

Best regards,

Alex Huffman

In order to include a statement about the importance of the atmospheric implication of our study, we extended the abstract by highlighting the need for ambient intercomparison studies for a qualified embedding of the INP variables in models. At the same time, to limit the abstract to 250 words, we shortened the abstract by excluding details about the INP methods and specific results about their intercomparability.

~~The formation of i~~ Ice crystal formation in mixed-phase 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. Models have shown that the presence of INPs in clouds can impact their radiative properties and induce precipitation formation. However, for a qualified implementation of INPs in models, it is needed that measurement techniques are able to accurately detect the temperature-dependent INP concentration. ~~INP concentrations can be detected online with high-time resolutions of minutes, or offline, where aerosols are collected on filters for hours to days.~~ Here we present measurements of INP concentrations in ambient air under conditions relevant to mixed-phase clouds from a total of ten INP methods over two weeks in October 2018 at the Puy de Dôme observatory in central France. ~~INP concentrations were detected in the immersion freezing mode, between ~ -5 °C and -30 °C. Two continuous flow diffusion chambers (CFDC; Colorado State University Continuous Flow Diffusion Chamber, CSU-CFDC; Spectrometer for Ice Nuclei, SPIN) and an expansion chamber (Portable Ice Nucleation Experiment, PINE) measured the INP concentration with a time resolution of several minutes and at temperatures below -20 °C. Seven offline freezing techniques determined the temperature-dependent INP concentration above ~ -30 °C using water suspensions of filter-collected particles sampled over 8 hours (FRankfurt Ice Nuclei Deposition FreezinG Experiment, FRIDGE; Ice Nucleation Droplet Array, INDA; Ice Nucleation Spectrometer of the Karlsruhe Institute of Technology, INSEKT; Ice Spectrometer, IS; Leipzig Ice Nucleation Array, LINA; LED based Ice Nucleation Detection Apparatus LINDA; Micro-Orifice Uniform Deposit Impactor–Droplet Freezing Technique, UNAM-MOUDI-DFT).~~ A special focus in this intercomparison campaign was placed on having overlapping sampling periods. Although quite different

measurement principles were used, the majority of the data are within a factor of 5, demonstrating the suitability of the instruments to derive model-relevant INP data.
Lower values of comparability are likely due to instrument-specific features such as aerosol lamina spreading in continuous-flow diffusion chambers~~, for the methods: INP concentrations measured with the online instruments were compared within 10 minutes and at the same temperature (±1 °C), while the filter collections for offline methods were started and stopped simultaneously and the obtained INP freezing spectra were compared at 1 °C steps. The majority of INP concentrations measured with PINE agreed well with the CSU-CFDC within a factor of two and five (71% and 100% of the data, respectively). There was a consistent observation of lower INP concentration with SPIN, and only 35% of the data are within a factor of two from the CSU-CFDC, but 80% of the data are still within a factor of five. This might have been caused by an incomplete exposure of all aerosol particles to water-supersaturated conditions within the instrument – a feature inherent to CFDC-style instruments –~~ demonstrating the need to account for ~~aerosol lamina spreading~~ such phenomena when interpreting INP concentration data from online instruments. ~~The comparison of the offline methodsrevealed that more than 45% of the data fall within a factor of two from the results obtained with INSEKT. Measurements using different filter materials and filter holders revealed no difference in the temperature-dependent INP concentration at overlapping temperatures. However,~~ Moreover, consistently higher INP concentrations were observed from aerosol filters collected on the rooftop at the Puy de Dôme station without the use of an aerosol inlet.~~, compared to measurements performed simultaneously behind the whole air inlet system~~

Moreover, the aspect of the atmospheric implication with regard to the precision in the INP measurement technique is very important and not discussed sufficiently in our manuscript. Therefore, we now add statements about different modeling studies, investigating the effect of a variability in the INP number concentration on cloud properties, to better assess the meaning of an inter-comparability of the instruments within certain factors.

We add to the introduction, lines 28 – 32:

The first formation of ice in mixed-phase clouds is triggered by specific aerosol particles, called ice-nucleating particles (INPs; Vali et al., 2015). 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) and related radiative properties (e.g., Vergara-Temprado et al., 2018). In addition, INPs have an impact on precipitation formation (e.g., Mülmenstädt et al., 2015; Field and Heymsfield, 2015; Fan et al., 2017). However, the identification and quantification of ambient INPs remain challenging due to their rarity (Kanji et al., 2017) and limitations in measurement techniques (DeMott et al., 2017; Cziczo et al., 2017). For a better integration of INPs in models, that is required to improve the representation of ice crystal formation and evolution in clouds (e.g., Coluzza et al., 2017; Burrows et al., 2022), a certain precision in INP measurement techniques is required, as studies have shown that a variability in the temperature-dependent INP number concentration impacts the representation of cloud properties (e.g., Phillips et al., 2003; Ervens et al., 2011; Tan et al., 2016; Vergara-Temprado et al., 2017; French et al., 2017).

,as well as to the end of the conclusion, lines 859 – 870:

For a better understanding of the formation and evolution of ice in clouds, it is essential to integrate observations of INPs in numerical models (e.g., Coluzza et al., 2017; Burrows et al., 2022). This requires a certain precision in INP measurement technique, as studies have shown that variations in the modeled INP concentration can lead to changes in the balance between ice and supercooled water, with a significant impact on the radiative properties of clouds and precipitation processes (e.g., Tan et al., 2016; Vergara-Temprado et al., 2017; French et al., 2017). Phillips et al. (2003) found a significant impact on modeled microphysical processes and precipitation rates when varying INP concentrations by factors of

10, indicating that observational errors need to be smaller than this factor. Ervens et al. (2011) investigated the partitioning between ice and water in Arctic mixed-phase clouds and only revealed an effect when INP concentrations were increased by a factor of 5. The results from this ambient intercomparison campaign revealed that the majority of the data from the different INP measurement techniques, in their original configuration, are within a factor of 5, which generally demonstrate their suitability to derive model-relevant INP data.

References

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