Author response to comments by Gabor Vali (Referee #1):

We thank Gabor Vali for his valuable comments and recommendations to our manuscript. We feel that due to these comments and our respective adjustments, the readability and quality of our manuscript is now improved, but intently anticipate reassessment by the referee.

In the following, we address the referee comments and describe adjustments made. For this, the referee comments are given in blue and our answers in black. When referencing page, line, and section numbers, we always refer to the first version of the manuscript unless otherwise stated.

The paper presents a newly designed sampling device for atmospheric ice nucleating particles (INPs). It is designed for use in research aircraft. The design is described in detail and test results are presented to show the sampling efficiency and reliability of the device. The significance of the results is the prospect that it will aid in obtaining research results on atmospheric INPs at higher spatial and temporal resolution than has been possible with other systems. This can be specially useful when coupled with cloud and precipitation studies. The paper addresses the relevant questions thoroughly, although with some limitations and with some superfluous material. The topic of the paper fits well within the scope of AMT.

The manuscript presents an evaluation of the HERA sampling unit specifically for INPs. To some extent this is justified as the stated goal for HERA is to collect samples for INP studies. On the other hand, it makes the paper somewhat convoluted and focus is lost on the fact that the novelty is the HERA sampling, not the filter processing. The latter issue has been extensively treated in previous publications. Some new data are included here on pore-size dependence but really that should be a topic examined on its own.

Except for rather special situations, INPs can be expected to be dynamically just like other aerosol particles. Thus, transmission and collection efficiencies for INPs of given sizes, densities and shapes can be expected to be the same as for similar particles of other kinds. In view of that, it would be better if the paper separated the HERA sampling efficiency from INP processing issues. In fact, it is curious why the authors focus only on INPs and do not leave open the use of HERA for aerosol sampling for other types of aerosol analyses.

As the referee points out, the focus of HERA is the sampling of INPs. This is why we chose to validate the collection efficiency via immersion freezing measurements. These involve a number of steps which are specific to the here presented measurements and the used setups and must be described in the manuscript. In order not to distract from the results, we shifted the specifics of the INP analysis to the Appendix, where it is easily available to the reader. The specifics of the sampling remain in the main part of the manuscript. With that, and some additional information concerning sampling efficiencies on aircraft and in the HERA-HALFBAC comparison experiments, we hope to have shifted the focus more towards filter sampling.

We chose the method of validating the collection efficiency of HERA via filter sampling and immersion freezing measurements of the filter washing water, since this is the general use case for HERA. The general aerosol particle population is usually analyzed with the help of online instruments on aircraft, which offer a higher temporal resolution. Of course, there are some exceptions to this. We added the following sentence in the introduction to better clarify the scope of the study: “While the HERA filter samples can be used for a number of different types of aerosol particle analyses, e.g., scanning electron microscopy for particle morphology analysis (Sanchez-Marroquin et al., 2021; Seifried et al., 2021) or ion chromatography for bulk chemical composition analysis (Kwiezinski et al., 2021), this study focuses on the application for immersion INP measurements.”

The paper also gets complicated by not clearly defining that installation-dependent other factors, specially inlet characteristics, are not subjects of this paper. Only in the discussion of the flight result (Section 4) do these issues have a place, as a specific instance of information needed to interpret the results.

Indeed, it was our original intention to separate the characterization of HERA itself from installation-dependent factors. However, we also understand the request for information on the transmission efficiency on aircraft, which was also brought forward by referee #2. We performed calculations of transmission efficiency for the CIRRUS-HL setup and added the results in Sec. 4: “[…] Assuming spherical particles with a density of 2 g/cm, an inline pressure of 340 mbar, and an inline temperature of 26 °C (corresponding to ~200 m/s TAS), D_{50} at the HASI is 2.7 µm (aspiration and transmission efficiency). The HALO-CVI sampling line to HERA had a total length of ~5 m (10 mm inner diameter until flow distribution at roughly half of total length, then contraction to 4.57 mm) resulting in D_{50} =2.2 µm for the above given conditions (D_{50} =4.7 µm from HALO-CVI inlet to flow distribution). ”

The HERA unit is aimed to facilitate the collection of filter samples for INP analyses by eliminating in-flight handling of the filters. This is a worthwhile goal, although controls used in many previous cases showed that manual exchanges of filters didn't produce noticeable contamination. The potential of HERA will be more fully evident once the operation is fully automated and the need for a technician attending to the collector is eliminated. Then, the remaining limitation will be the number of filters per flight to six. Selection of six truly meaningful sampling intervals will require difficult decisions by the flight directors. The paper could be more realistic about these issues. Also, it should be clarified if filter holders could be exchanged during flights or not. Perhaps this is already in the paper and was missed in reading it.

The referee is correct in mentioning that results from previous aircraft filter sampling do not report noticeable contamination. However, only very few studies mention the collection of field blanks in the first place, and those that do, did only collect them sporadically. Hence, contamination might not have been detected. To clarify this issue, the respective sentence in the original manuscript was changed to: “While no significant contamination was reported in the above mentioned studies, blanks were not taken during every flight and contamination might have been missed depending on the frequency of occurrence. ”

In principle, it would be possible to remove the filter holder inset in flight and replace the filter holders with “fresh” ones. This was added to the manuscript in Sec. 2.3. However, this is not possible on the HALO aircraft, where instrument parts must not be removed in-flight due to safety regulations.

It is correct, that the selection of meaningful sampling intervals is challenging. However, this challenge would prevail even with more available filter holders. To clarify the difficulties, we reformulated the paragraph in question in Sec. 2.3: “Any kind of aircraft INP filter sampling involves careful planning to achieve truly meaningful sampling intervals. In general, the flight pattern should be accounting for sampling periods under somewhat constant atmospheric conditions […]. In practice, the sampling strategy often must be adapted in-flight due to unforeseen changes in weather conditions and/or flight track. Consequently, fast decisions by the on-board operators are needed which can be easily realized with HERA due to the quick (< 30 s) and remote-controlled switching between filters. The number of six filters per flight was based on typical flight durations and expected INP concentrations in the free troposphere and so far was found appropriate in practice. If more filters are needed and the aircraft certification regulations allow for it, the filter holder inset could be removed in-flight and filter holders could be exchanged.”

The design of HERA is described in great detail. Yet, one misses information about overall size, weight and power requirements. Those are important factors for aircraft deployment.

We apologize for not mentioning these important facts. We included the following in Sec. 2.1: “The sampling unit measures 49 cm × 52 cm × 27 cm (width × depth × height), fits into a standard 19 inch rack unit, and weighs 22 kg. […] The pump unit measures 49 cm × 25 cm × 18 cm, weighs 13 kg […]. At full pump speed, the power consumption of HERA is ~400 W .”

Sampling efficiency is presented in terms of theoretical calculations. It would have been good to have support for the computed efficiencies from actual tests. The tests with Snomax are in that direction but for smaller sizes than the calculations (Fig. 2).

We agree with the referee. Sampling of supermicron particles was considered for the experiments with SNOMAX and ATD, but not performed due to the difficulties of supermicron particle generation and size-selection and the related significant increase in sampling times. For the submicron particles, our results fit the calculations of the transmission efficiencies. For now, nothing can be said about the actual transmission of supermicron particles. We added the following in Sec. 3.2: “Due to the particle generation setup, the sampling experiments were restricted to particles with mobility diameters ≤ 800 nm, where particle losses should be minimal according to the theoretical calculations (see Fig. 2, minimum transmission efficiency of 96.4 % for 800 nm particles sampled at a flow rate of 100 L/min and near-standard pressure). Regarding supermicron particles, a decrease in transmission efficiency is expected according to the calculations, but experimental results cannot yet be provided.”

A design criteria of 0.5 µm lower limit for particle sizes is given and transmission efficiencies are calculated for size above that limit. Small particles are less subject to losses. This is fine, but the justification given deserves some comments. Two references are cited in support of the decision for INPs. Both references show correlations of INP concentrations with particle sizes >0.5 µm but also show temperature dependence and variations with ambient conditions. None of the correlations are strong at higher nucleation temperatures and, in any case, correlations can arise from source or transport similarities without the INPs actually being >0.5 µm. Both studies were made with samples at ground level and there are other limitation as well in the two references. It is out of place for this manuscript to review the literature about INP sizes. It should however not give the appearance of a firm justification for the choice. Results obtained with the use of the HERA will have to be evaluated with the size question in mind, specially since the impact of INP sizes is also an issue for INP extraction and detection with the filter method.

The particle size range of > 0.5 µm was merely cited from the literature because correlation between the concentration of these particles and INP concentrations was observed. It was not our intention to communicate 0.5 µm as the lower size limit for particle collection with HERA. Transmission efficiencies were also calculated for particles < 0.5 µm (see Fig. 2). We agree that stating a lower size limit for INPs is not appropriate and decided to leave out any size information in this part of the manuscript to avoid confusion. The text was adapted in the following way: “ While there are several factors influencing the potential of an aerosol particle to act as an INP, size seems to be an important one, as the concentration of large particles has been shown to correlate with the INP concentration (Pruppacher and Klett, 1997; DeMott et al., 2010). INP sampling should hence be setup so that losses of large particles are minimized.”

With the foregoing in mind, it is recommended that the Introduction focus on the sampling issues from aircraft, that Section 3.1 be re-written to focus on showing the lack of sensitivity to flow-rate for small particles (as expected) for one filter setup. Sections 3.2, 3.3 and 4 are the main results to present. The use of 'deposition efficiency' should be avoided and it is easily confused with deposition nucleation tests. The discussion about cell fragments versus protein aggregates isn't effective because of the differences in physical dimensions; the sources of differences are more likely to be a question of sample aging and other treatment differences. The observed drop-off in detection at the higher temperatures is concerning but not a point to be treated in this paper.

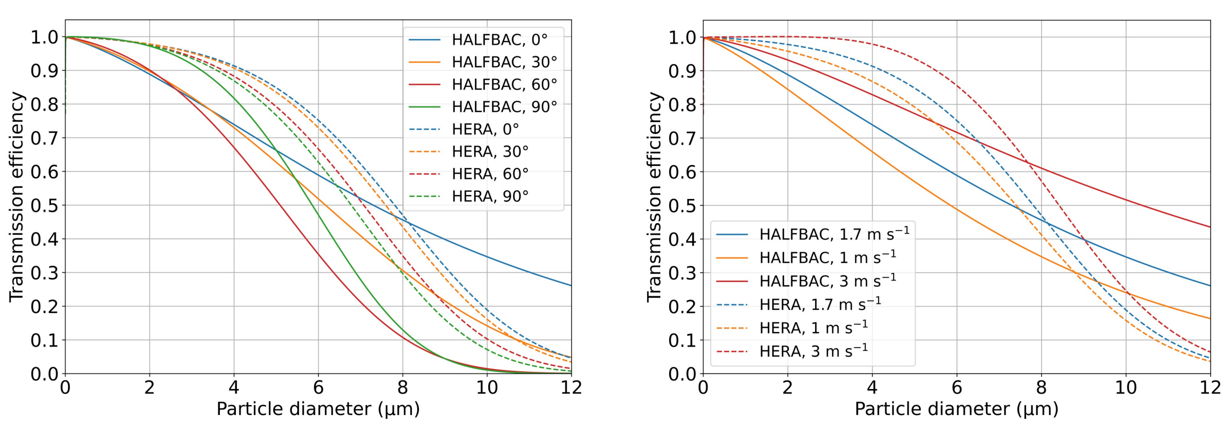
We are not completely sure that we understand the comment concerning the re-writing of the Introduction and Sec. 3.1. The introduction already includes an overview of INP sampling activities and issues on aircraft (p. 3-3, l. 80-101). Section 3.1 deals with the effect of filter pore size on INP sampling at a single flow rate. This experiment did not only focus on “small” particles, but was performed with polydisperse ATD and ambient air, which most definitely included some supermicron particles. We respectfully ask the referee to clarify his request concerning the content of the mentioned sections and we will implement adjustments in the next round of reviews.

The term “deposition efficiency” was replaced with “collection efficiency” at all instances.

Concerning the missing class A mode in the INP spectra of SNOMAX after filter sampling, we believe that our original explanation is a possible cause. The class A protein aggregates can be several tens of nanometers in size (Govindarajan and Lindow, 1988) and might very well be separated when the much larger *P. syringae* cells are fragmented in the atomizer. Wex et al. (2015) report sizes of cell fragments ranging from 50 nm to 250 nm after spraying with an atomizer. From our point of view, this argumentation is quite convincing and we would like to stick with it in the manuscript. However, we adjusted the paragraph slightly to leave more room for interpretation: "Interestingly, we do not observe the freezing mode above -5 °C [...] which can have several causes. This mode is commonly associated with the occurrence of large aggregates of ice nucleation active proteins which are found in the outer membranes of the P. syringae bacteria (Lindow, 1995; Schmid et al., 1997). Bacterial cells have been shown to be broken up into fragments when spraying a SNOMAX® suspension with an atomizer (Wex et al., 2015), reducing the probability of large protein aggregates being deposited on the filters. Another reason for the missing high-temperature mode could be the prolonged storage of the SNOMAX® batch leading to some kind of deactivation of the large protein complexes."

In Section 3.3. the experimental setup would be of interest and the reason for the wind-speed dependence more understandable. This is more to the central point of the paper (sampling efficiency).

We added more detail to the description of the experimental setup in Sec. 3.3, e.g., the inlet inner diameters of the two instruments. In accordance with comments by referee #2, we performed additional calculations of the aspiration efficiency in dependence of wind speed and direction to estimate the effect of these two quantities on the overall sampling efficiency. It turns out that in our experiments due to the smaller inlet diameter of HALFBAC (½ inch) in comparison to HERA (¾ inch), the HALFBAC collection efficiency is quite strongly influenced by changes in wind speed and direction (see figures below).



The left plot shows the sampling efficiency at a constant wind speed of 1.7 m/s (same as mean wind speed during sampling periods 4 and 5) with variable aspiration angle of 0° (inlet facing wind directly) to 90° (inlet facing wind at 90°). Solid lines represent HALFBAC (½ inch inlet), dashed lines HERA (¾ inch inlet). The right plot shows the sampling efficiency at a constant aspiration angle of 0° with variable wind speed ranging from 1 m/s to 3 m/s. In general, HERA samples particles with diameters ranging from 0 to ~8 µm, which comprises the vast majority of the urban background aerosol particle population (see measurements by Mordas et al., 2015), more efficiently than HALFBAC for the given parameters. It can be seen that an increase in aspiration angle and a decrease in wind speed cause particles to be sampled less efficiently with HALFBAC. At constant wind speed, D_{50} shifts from 7.3 µm to 5 µm with a change in aspiration angle from 0° to 60° (see left plot). If the inlet is facing into the wind (aspiration angle = 0°), D_{50} is shifted from 10.4 µm (3 m/s) to 5.8 µm (1 m/s). The sampling periods with the largest discrepancies in INP concentration between HERA and HALFBAC (3, 4, and 5) are also the ones with the lowest wind speed and the strongest variability in wind direction, and could thus be the periods with the least efficient sampling with HALFBAC. This information is now included in Sec. 3.3.

In summary, this is paper about an important new instrument and its characterization for INP sampling. Much good material is presented. Improvements can be gained by simplification of the paper.

References (only those not included in original manuscript reference list):

Govindarajan, A. G., & Lindow, S. E. (1988). Size of bacterial ice-nucleation sites measured in situ by radiation inactivation analysis. *Proceedings of the National Academy of Sciences*, *85*(5), 1334-1338.

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Pruppacher, H. R. and Klett, J. D.: Microphysics of Clouds and Precipitation, Kluwer Academic Publishers, Dodrecht, The Netherlands., 1997.