Author response to RC1

December 22, 2024

We are grateful for the helpful comments and suggestions from the reviewer. Below the reviewers comments are in blue with our responses in black directly below.

The manuscript describes the application of a newly deployed mobile filter sampling system, operating on an uncrewed aerial vehicle (UAV). The development and improvement of the sampling technique over three subsequent measurement campaigns is well documented. Also, the text is well written, everything is well described.

However, occasionally statements need correction, and in some parts additional clarifications are needed, as specified in the general comments. Also, there is one mayor criticism I have that may well go beyond this study: For testing new equipment, test flights were done in northern Finland. It is not clear why they were not done at the home institute, and instead a lot of travel and transport of both people and equipment was done to get these results. This does not diminish the quality of this work, but should at least be considered in future developmental work.

After below comments will have been addressed, I can support publication in AMT.

The authors agree that the reduction in travel and transport of both people and equipment should be a common goal for developmental work, especially in regard to climate change. We do see this as a valid criticism and want to offer some answers on why we choose the area in northern Finland.

We have a long standing collaboration with the Finnish Institute of Meteorology (FMI), having participated in previous Pallas Cloud Experiment (PaCE) campaigns. Their experience in operating UAVs was highly beneficial for the success of the development of our filter-based INP sampling setup.

Regulations for the use of UAVs are very restrictive in Germany and in general in Europe. This is one major hurdle for the operation of UAVs. The existence of a temporary danger zone around the Sammaltunturi station in Finland allows a lot of flexibility. The collaboration with the FMI and their offer to use this temporary danger zone for our testing and validation experiments was highly beneficial and added to the overall success of this project.

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1 General comments

Line 69: "regions with low aerosol concentrations, such as northern Finland" sounds wrong. An annual cycle of the Arctic aerosol has been understood for some time now. Concerning size distributions or number concentrations, you can check out e.g. Tunved et al. (2013) or Freud et al. (2017). Arctic haze contributes particles in winter time, new particle formation causes elevated particle concentrations in summer time. Concerning INP concentrations, it has also been understood that there is an annual cycle with high concentrations possible already in spring and advancing well into autumn (see e.g., Creamean et al., 2018, Wex et al., 2019; Porter et al., 2023; Schneider et al., 2021, with the latter one from your institute). Therefore, the assumption of low INP concentrations in norther Finland is somewhat amazing and wrong. Please correct. (BTW: Check your literature list. For Schneider et al. (2021), I saw that the doi refers to the discussion paper. – I did not check the rest.)

We thank the reviewer for this comment. We agree that our statement is too strong and not precise enough. Our campaigns were either conducted during spring or autumn near the Sammaltunturi Global Atmosphere Watch (GAW) site (67°58'N 24°7'E, 565 m above sea level (asl), northern Finland, Lohila et al. 2015). Sammaltunturi is located in a subarctic environment above the tree line around 180 km north the Arctic circle. While the total number concentration at Sammaltunturi can reach higher concentrations ($< 10\,000\,\mathrm{cm}^{-3}$), this is as the reviewer states only the case for summertime. Spring and autumn concentrations are typically one order of magnitude less, see for example Lohila et al. 2015.

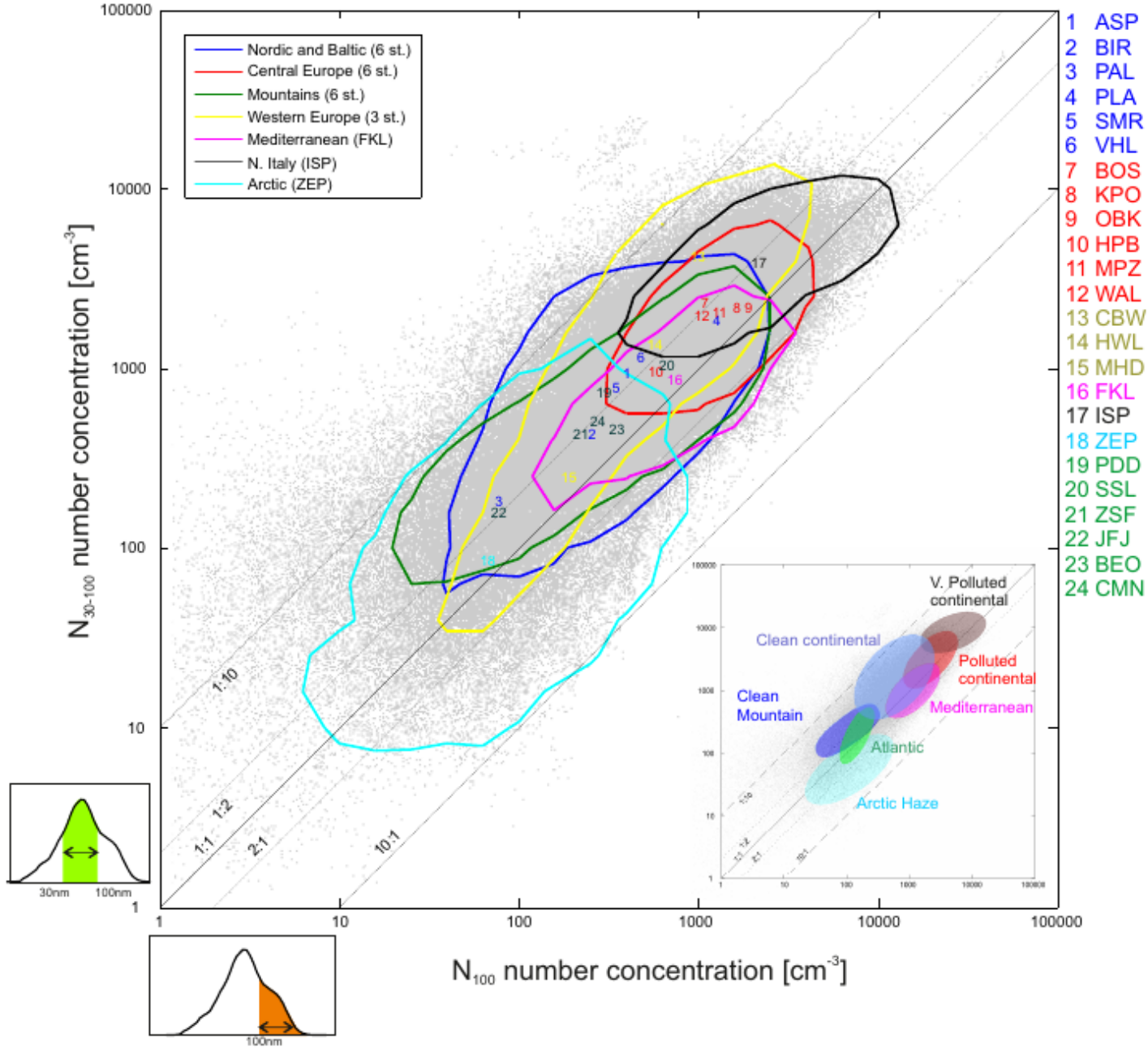

Figure 1: Figure 13 from Asmi et al. 2011.

We also want to add that while Arctic haze does contribute during winter and spring time in the Arctic, the area around Sammaltunturi is typically considered subarctic and to the authors knowledge is generally not representative for Arctic haze compared to stations like Zeppelin that are typically inside the polar dome during winter and spring (e.g., Asmi et al. 2011, see fig. 13 therein and fig. 1). Tunved et al. 2013 describe the aerosol measurements from the Zeppelin station, which is very much representative for an Arctic station that is affected by Arctic haze. Freud et al. 2017 reports measurements "from five sites around the Arctic Ocean (Alert, Villum Research Station – Station Nord, Zeppelin, Tiksi and Barrow)" but not from Sammaltunturi itself. Schmale et al. 2022 analyzed aerosol measurements of 10 Arctic observatories, one of them being Sammaltunturi (denoted as Pallas). Sammaltunturi is much more influenced by anthropogenic emissions from Europe and Asia and by biogenic emissions from the boreal forest compared to other Arctic stations, such a Zeppelin (Schmale et al. 2022).

Asmi et al. 2011 compares different stations in mainland Europe and subarctic and Arctic locations on aerosol concentration for a two-year-period. Table 4 of Asmi et al. 2011 shows the mean number concentration of Sammaltunturi (denoted as Pallas) as 198 cm^{-3}, 387 cm^{-3} and 211 cm^{-3} for the size ranges 30 nm to 50 nm, >50 nm and >100 nm, respectively. Only two stations report lower concentrations, them being a high-altitude station (Jungfraujoch in the Swiss Alps) and a high Arctic station (Zeppelin on Svalbard). This is even more pronounced when looking at the lower quantiles. We therefore think our original statement is not wrong. We agree that we need to be more careful with this statement and have therefore added the following to the mentioned sentence:

old This setup proved the technical feasibility of measuring INPs with sample times between 45 and 90 minutes in regions with low aerosol concentrations, such as northern Finland.

new This setup proved the technical feasibility of measuring INPs with sample times between 45 and 90 minutes in regions with low aerosol concentrations, such as the area around Sammaltunturi in northern Finland during the measurement periods in spring and autumn.

In reference to INP measurements conducted by our institute in southern Finland (Hyytiälä, Smear II, Schneider et al. 2021), we would like to add that the measurement location is quite different to Sammaltunturi and that the mean number concentration in Hyytiälä (Smear II, southern Finland) is higher compared to Sammaltunturi: $345\,\mathrm{cm}^{-3}$, $1053\,\mathrm{cm}^{-3}$ and $450\,\mathrm{cm}^{-3}$ for the same size ranges (Asmi et al. 2011). This results in factors of 1.74, 2.72 and 2.13, respectively. If we consider a similar ice-active fraction of the aerosols, this would equal that INP concentrations measured at Sammaltunturi are lower by a similar factor.

To conclude: we do not make the statement that Sammaltunturi generally features low INP concentrations, but we understand that it might be implied from our wording. We have therefore added an additional sentence:

new While low aerosol concentrations might also lead to lower INP concentrations, this is not always true, since some aerosols might be much more ice active than others and therefore contribute disproportionally to the INP concentration (e.g., Hoose and Möhler 2012).

We are grateful to the reviewer for reviewing the literature and have changed the reference of Schneider et al. 2021 to accurately refer to the finalized publication. We have also checked the rest of the literature to make sure there are no other mistakes to the best of our knowledge.

Line 134-135: Why do you subtract the water background, but not, instead, the background from the field handling blank? By this, you create some in-between state with the background partially, but not fully, subtracted.

To the authors' knowledge no consensus has been reached in the INP community on how to handle handling blank filters. We have discussed if we would subtract the handling blank from the filter, but have ultimately decided not to, because of the following reasons: the water is subtracted for the frozen fraction, since one is unable to associate it with any amount of air volume sampled. The handling blank is analysed in the same way as the actual filter samples and the handling blank is typically very close to its water background. This introduces issues since the water background is slightly different for each experiment. Therefore, it is also slightly different for the handling blank. For these reasons, we use the handling blank to assess our handling procedure, but do not subtract its frozen fraction from the original samples. This way we can flag filters as invalid, if they were taken after a handling blank filter whose aerosol suspension shows a contamination.

Line 169: Why are you talking about campaign 2 here at the beginning of the description of campaign 1? Did you not take field handling blanks during campaign 1? Then better mention that explicitly.

We do not report any results from campaign 1. Campaign 1 was a proof-of-concept study and meant to test the technical feasibility of the general setup. One major objective of campaign 1 was the investigation of the INP concentration obtained from such a filter-based UAV setup with relatively short sampling times with a focus on area with typically and relatively low aerosol concentrations. For campaign 2 we report more substantial and significant results, where we also focused on taking handling blanks.

We would like to show some results from campaign 1 to the reviewer (see fig. 2), showcasing that we did detect INPs with the first setup, but we would like to focus our results on the second setup:

Line 174 and Fig. 6: Do you have any idea why the handling blank on the UAV is higher than that on ground? And is it right that there was only one handling blank for UAV and one for the ground? Then please mention this explicitly.

There is no identified reason why the handling blank on the UAV is higher than on the ground. The frozen fraction of the handling blank for the UAV is still below the frozen fraction of the UAV filters. Unfortunately we only took one handling blank filter for this setup. We have changed the following sentence to mention is explicitly:

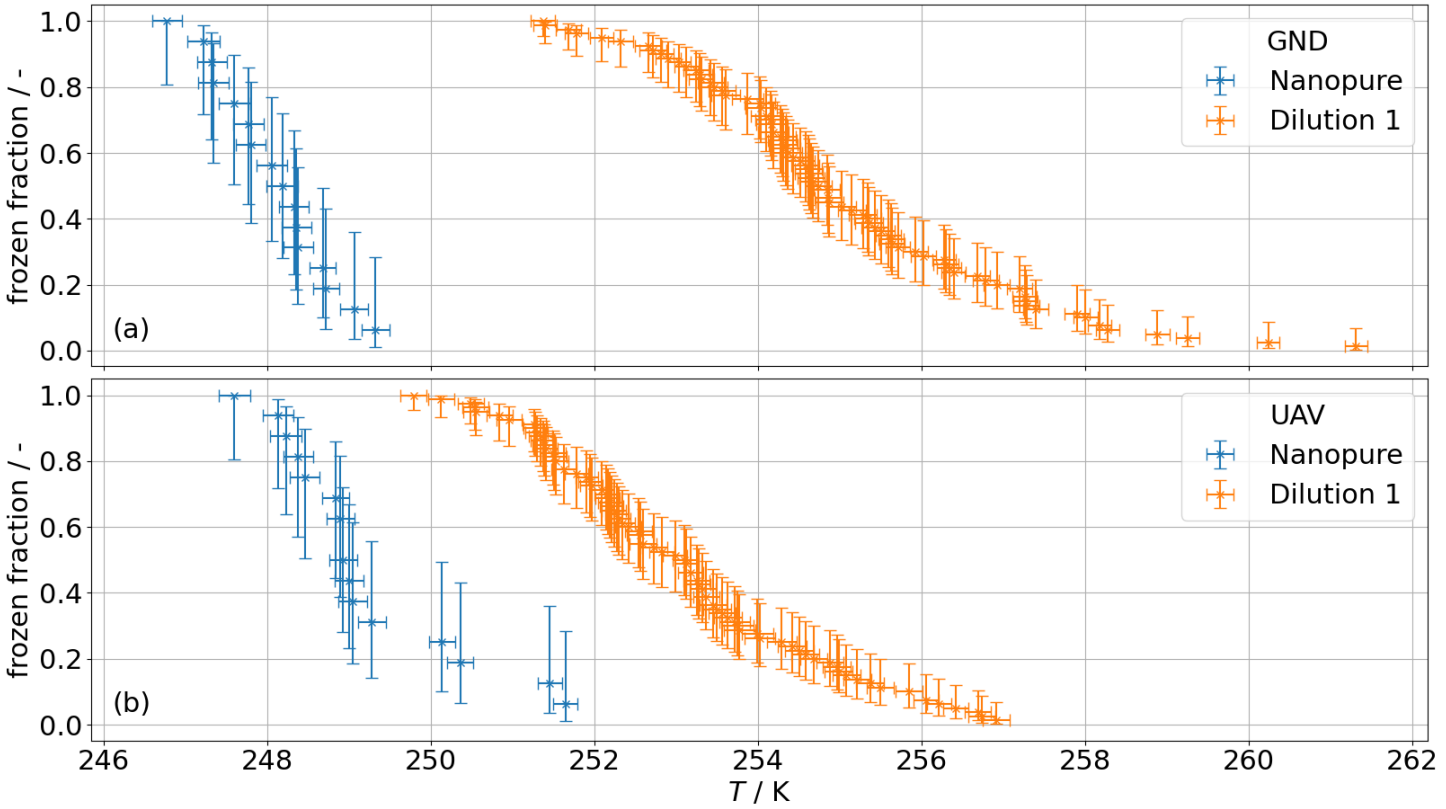

Figure 2: Frozen fraction of an aerosol sample collected during campaign 1 on the ground (panel (a)) and on-board of a UAV (panel (b)). Both suspensions show a clear separation from the water background (denoted as Nanopure).

old To demonstrate the scientific feasibility, *the inverse of the liquid fraction,* the frozen fraction, of the UAV and ground filters are compared to their respective handling blank filters taken during campaign 2.

new To demonstrate the scientific feasibility, the frozen fraction of the UAV and ground filters are compared to their respective handling blank filters taken during campaign 2. Only one handling blank filter was taken for each setup.

Note that the part in *italic* was changed due to comments from reviewer 2.

Line 146 ff: About the detection limit: The argument in this paragraph is too complicated and can be simplified a lot. The complication is by first assuming that in the whole batch there should be one INP, then getting a value for that, but then saying (correctly), that it is not the whole batch that is examined, but only one droplet. The number given in the text for the whole batch is useless, and no number (or range) is given here for the results of equation (3), which really is the lower detection limit. (I only later on found one value in line 214.) Instead, it could just be said that the detection limit comes from equation (2) with the assumption of one frozen droplet (i.e., one of the 64 droplets frozen, or f_l = 63/64 = 0.984). That will be the minimum value you get as a concentration, and is in the same order of magnitude than the value you give in line 214. Modify / simplify this paragraph!

We are thankful for the careful reading of this section from the reviewer. The reviewer is discussing the resolution of the instrument, which is of course correct in the sense that our minimal resolution is directly tied to the amount of simulated droplet. Our assumption is related to the total volume of the water used to wash the aerosol of the filter. There are 64 wells filled with 50 µL each, which equals a total of 3200 µL = 3.2×10^{-3} L. Our assumption is now that the experiment does detect an INP if there is one INP in any of those wells. This signal would be different to the pure water signal. Combining this with a volume of the washing water of 5×10^{-3} L, results in a factor of roughly 1.56. The actual detection limit is therefore by this factor higher than the simple assumption of the inverse of the sampled air volume. We update this paragraph to make our intention more clear:

old The lower detection limit can be estimated by the condition that a single INP has to exist to initiate freezing, i.e. for a sampled volume of 500 $\mathrm{L}_{\mathrm{std}}^{-1}$, the rough estimate for the lower detection limit for the INP concentration is $c^*_{\mathrm{INP,low}} = 2 \times 10^{-3}\,\mathrm{L}_{\mathrm{std}}^{-1}$. This detection limit also depends on the analysis, since the whole suspension is not used for one analysis.

new The lower detection limit can be estimated by the condition that a single INP has to exist to initiate freezing, i.e. for a sampled volume of $V = 500\,\mathrm{L}_{\mathrm{std}}^{-1}$, the rough estimate for the lower detection limit for the INP concentration is $c^*_{\mathrm{INP,low}} = V^{-1} = 2 \times 10^{-3}\,\mathrm{L}_{\mathrm{std}}^{-1}$. This detection limit also depends on the analysis, since only a fraction of the whole suspension is used for one analysis.

Figure 7: In the caption, you refer to the steepness of the sampling curve such that the impression arises that the steepness of the curve depends on the sampling time. But such a dependency does not exist! The steepness of the curve reflects which INPs are there in which concentrations. Each INP has its characteristic freezing (or ice-activation) temperature (in the non-stochastic approach you are using here). And the curve shows how many INPs are there that are ice-active at the different temperature. So the shape of the curve has nothing to do with the sampling time. You can only argue, as you do, that by increasing the volume of sampled air you also increase the maximum temperature at which you get data (by decreasing the minimum detection limit), and that you may see different parts of the curve. But the curve at a certain temperature range will be the same, independent how long you sample. Otherwise the whole measurement approach would not make sense. And that increase in maximum temperature is not much, as doubling the sampling time only halves concentration, which is not much gain in terms of temperature at which data is available.

Bottom line: Modify the caption of Fig. 7 (and elsewhere in the text if needed) such that this wrong impression of a connection between sampling time and steepness of the curve does not appear any more.

We thank the reviewer for their careful reading. We did not want to raise the impression that the steepness of the curve is associated with the sampling time. We removed the discussion of the steeper curve and modified the original sentence to remove any ambiguity:

old It can be seen that lower INP concentrations can be detected, as well as a steeper freezing curve.

new It can be seen that lower INP concentrations can be detected due to the increased sampling time.

Line 187: Again, as above, the Arctic is not on its own a region with clean air (see e.g., Arctic haze) or low INP concentrations (see my remark for line 69). Please reword this sentence.

See our answer to your previous comment. We have moved this sentence into the Conclusion and Outlook section and added an additional part to explain why the Arctic is especially interesting for measuring the vertical distribution of INPs:

new This is the case especially in the Arctic due to its characteristically stratified atmosphere (Graversen et al. 2008).

Line 191: "This difference highlights the importance of measuring the vertical distribution." While I agree that differences in INP concentrations are to be expected with altitude, it becomes less clear in the presented data than what I would have expected. It would be more correct to state that while you do not present data showing big differences between ground and UVA, stronger differences can be expected and measuring the vertical distribution is important.

We want to highlight that this setup is able to measure INP concentrations at different altitudes and is therefore able to detect differences, but based on the data presented in this paper no conclusions about potential differences can be drawn. We have adjusted the following sentence:

old This difference highlights the importance of measuring the vertical distribution of INPs to evaluate their influence on cloud microphysics, which we demonstrate is possible with the UAV sampling system described herein.

new This difference, albeit small, highlights the importance of measuring the vertical distribution of INPs to evaluate their influence on cloud microphysics. The data presented in this study demonstrate that such investigation is possible with the UAV sampling system described herein.

We also would like to add that we did additional and more comprehensive measurements during a Pallas Cloud Experiment (PaCE) campaign in autumn 2022, which will be published in the future.

Line 203: Referring to Chapter 4 and the overall work:
Given that we are living in times of climate change and should rather watch our resources, a question arises, overall: Why were these tests not done at home, but instead, for a few research fights, a lot of travel and transport was done?

We agree with the reviewer and would like to refer to our answer at the beginning of this response. Each and every travel should be carefully evaluated and a decision should be made with the consequences and the benefits evaluated against each other.

Also: How did you realize that the different inlets performed differently? Only by modeling? Please add this information to the text.

We have only used well-established theoretical equations for the estimation of the transport efficiency, which can be found in standard text books (e.g. Kulkarni 2011). To reflect this better in the text, we changed the following sentence:

old This change enhanced the sampling efficiency of larger aerosol particles due to fewer bends...

new This change enhanced the theoretical sampling efficiency of larger aerosol particles due to fewer bends...

Line 215: You claim that INP concentrations are "usually showing a decrease of INP concentrations at higher altitudes". This cannot be seen from the data in Fig. 7, given the agreement within measurement uncertainty in both panels and the fact that data obtained from the UAV is even higher than data collected at ground in the left panel. Either show different data in Fig. 7 if you have them, or correct the text (here and throughout the manuscript) accordingly.

We have adjusted this sentence from the manuscript. For these first results, we do see a general trend, but we agree that we need a larger amount of data to create a reliable statistic and give concrete conclusions. We changed the earlier part:

old Some flights show a difference between the UAV filter compared to a concurrent ground-based filter, usually showing a decrease of INP concentrations at higher altitudes.

new Some flights show a difference between the UAV filter compared to a concurrent ground-based filter, but based on the current dataset it is not possible to draw a statistically relevant conclusion.

Line 218-222: In this part, you give future plans instead of highlighting why your approach was a useful addition. The impression arises that you found that your new approach is not very good. You can leave that as it is, but maybe it diminishes the value of your work too much.

We have adjusted this sentence:

old Future improvements will include size distribution measurements in addition to INP measurements via small, lightweight optical particle counters.

new Future experiments will include size distribution measurements in addition to INP measurements via small, lightweight optical particle counters.

And also added two new sentence to the previous paragraph to better highlight the usefulness of our approach:

old

new The current work provides a solid foundation for understanding INP concentrations in varied atmospheric conditions. Additional measurements will further enhance the statistical robustness and reliability of our findings.

Line 223: You can already use backwards trajectories for your own UAV measurements, so the question arises why you did not do that if you deem them so valuable, here.

We hope to combine different approaches in the future for identifying air mass and aerosol origin. The focus of this study is on the technical aspect of the measuremeent setup, and gives an overview of the improvements made to refine our method. We aim at including more information about aerosol characteristics and origin, e.g., by using FLEXPART (Flexible Particle Dispersion Model, Pisso et al. 2019) in future publications. This is one major aspect of a field campaign that we participated in during autumn 2022 (Pallas Cloud Experiment 2022) which will be published soon.

Line 224: Why especially in the Arctic? Please justify (half a sentence can be enough) if you want to make this claim.

We removed this part of the sentence and added it as a new sentence with a justification:

old A vertical distribution of INP concentrations, especially in the Arctic, could also be helpful to validate as well as complement models to connect ground- and aircraft-based measurements.

new A vertical distribution of INP concentrations could also be helpful to validate as well as complement models to connect ground- and aircraft-based measurements. This is the case especially in the Arctic due to its characteristically stratified atmosphere (Graversen et al. 2008).

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2 Specific and editorial comments

Line 3: Add "in Finland" to the site description.

We added this part.

old A mobile sampler for collecting aerosol particles on an uncrewed aerial vehicle (UAV) was developed and tested during three consecutive Pallas Cloud Experiment campaigns in the vicinity of the Sammaltunturi Global Atmosphere Watch site (67°58' N, 24°7' E, 565 m above sea level).

new A mobile sampler for collecting aerosol particles on an uncrewed aerial vehicle (UAV) was developed and tested during three consecutive Pallas Cloud Experiment campaigns in the vicinity of the Sammaltunturi Global Atmosphere Watch site (67°58' N, 24°7' E, 565 m above sea level) in Finland.

Line 26-28: This sentence is totally correct, but not needed in the context of this work. Consider deleting it, although I will not force you to do it.

We would like to keep this part in as an additional motivation of our research.

Line 111: wrong format for citing Schneider et al. (2020). Try using citep[text preceeding citation][text following citation]label. (If this won't work for AMT, there will be a similar way, mayby with <...> instead of [...]).

We are assuming the reviewer means that we should remove the brackets around the year of the citation. We changed the original command from
`INSEKT, see e.g.~\citet{Schneider2020}`
to
`\citep[INSEKT, see e.g.,]{}{Schneider2021}`, which results in:

old Schneider et al. (2020)

new Schneider et al. 2021

We have also adjusted this for the following citations to be consistent:

old The newly developed UAV-based aerosol sampler was used and further developed during three Pallas Cloud Experiment campaigns, close to the Sammaltunturi Global Atmosphere Watch (GAW) site (67°58'N 24°7'E, 565 m above sea level (asl), northern Finland, Lohila et al. (2015)), which took place during autumn 2020, spring 2021, and autumn 2021.

new The newly developed UAV-based aerosol sampler was used and further developed during three Pallas Cloud Experiment campaigns, close to the Sammaltunturi Global Atmosphere Watch (GAW) site (67°58'N 24°7'E, 565 m above sea level (asl), northern Finland, Lohila et al. 2015), which took place during autumn 2020, spring 2021, and autumn 2021.

old The filter-based setup was used and further improved during three field campaigns to collect INPs in low aerosol concentration environments (i.e., northern Finland, Lohila et al. (2015)) at different heights up to 1 km agl.

new The filter-based setup was used and further improved during three field campaigns to collect INPs in low aerosol concentration environments (i.e., northern Finland, Lohila et al. 2015) at different heights up to 1 km agl.

Line 183-184: Which weight are you talking about, here? And is this important?

We are calculating the flow with a weighted mean of the two flights. The weight here is the sampling time and not associated with any physical weight. We have added the following statement to make this clearer:

old The flow over the filter is calculated by the mean pressures during sampling, whereas the weight is defined by the sampling time for each flight.

new The flow over the filter is calculated by the mean pressures during sampling. The actual flow is the weighted arithmetic mean, where the weight is defined by the sampling time for each flight.

Line 187: Names are always capitalized -> "Arctic".

We have changed this.

old This is especially important in clean-air environments such as the arctic (Bigg 1996; Hatakka et al. 2003; Lohila et al. 2015; Šantl-Temkiv et al. 2019).

new This is especially important in clean-air environments such as the Arctic (Bigg 1996; Hatakka et al. 2003; Lohila et al. 2015; Šantl-Temkiv et al. 2019).

Due to a comment from the second reviewer, we have also moved this sentence to the Conclusion and Outlook section.

Equation A2: Explicitly state also here, that Ddn is a specific dilution.

We have added this statement and made a reference to equation A7.

old This results in the following formula for the variance:

$$\mathrm{Var}(c_{\mathrm{INP,air}}) = c_{\mathrm{INP,air}}^2 \left[\left(\frac{\Delta V_{\mathrm{sol}}}{V_{\mathrm{sol}}} \right)^2 + \left(\frac{\Delta V_{\mathrm{well}}}{V_{\mathrm{well}}} \right)^2 + \left(\frac{\Delta V_{\mathrm{air}}}{V_{\mathrm{air}}} \right)^2 + \left(\frac{\Delta d_n}{d_n} \right)^2 \right] . \tag{A2}$$

new This results in the following formula for the variance:

$$\mathrm{Var}(c_{\mathrm{INP,air}}) = c_{\mathrm{INP,air}}^2 \left[\left(\frac{\Delta V_{\mathrm{sol}}}{V_{\mathrm{sol}}} \right)^2 + \left(\frac{\Delta V_{\mathrm{well}}}{V_{\mathrm{well}}} \right)^2 + \left(\frac{\Delta V_{\mathrm{air}}}{V_{\mathrm{air}}} \right)^2 + \left(\frac{\Delta d_n}{d_n} \right)^2 \right] , \tag{A2}$$

where d_n denotes a specific dilution (see Eq. (A7)).

Line 265: As this part is quite separate from the main text, specify here which flow meter you are referring to here, with "the flow meter".

We have added this information in the Appendix to make it clear which flow meter we are referring to:

old The maximal values of this sinus are higher than the rated full scale of the flow meter.

new The maximal values of this sinus are higher than the rated full scale of the flow meter (SFM4100).

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