Response to Reviewers

We are very thankful to the reviewers for their comments and suggestions, which have substantially improved the quality of the manuscript. Each comment is discussed separately with the following typesetting:

Reviewer's comments

Authors response

"Text from the revised version of the manuscript"

Changes in the manuscript due to comments from Reviewers

Reviewer #1

This manuscript presents the development and validation of the GRAnada Ice Nuclei Spectrometer (GRAINS) and uses it to compare different filter substrates and extraction methods for offline INP analysis. The question of how sampling substrates and extraction procedures influence INP quantification is highly relevant to the community. In particular, the assessment of quartz filters is important and potentially valuable, as they are widely used in routine aerosol monitoring networks but rarely applied in INP studies. Identifying and validating new sampling substrates could have far-reaching implications for the INP community, where polycarbonate filters have long been the standard choice. Because of the potential impact, it is essential to examine the results with exceptional care. Overall, the study fits well within the scope of AMT, and it could become suitable for publication after the following comments are addressed.

Major Comments:

A substantial portion of the manuscript describing the instrument characterization and intercomparison is well written and technically solid. My main concerns focus on the key scientific question of the paper. Can quartz filters replace polycarbonate filters for offline INP sampling? Under what conditions would such a substitution be valid? And how should the two quartz-based extraction methods (quartz punch and punch-washed) be interpreted relative to each other? Unfortunately, in its current form, the presented dataset does not convincingly answer these questions. Several interpretations appear speculative or insufficiently supported by data or physical theory. I strongly encourage the authors to revisit these sections, refine the language, introduce clear limitations and uncertainties, and tone down the strength of the conclusions.

We thank the reviewer for the overall review. We would like to highlight that our goal is not to state that quartz filters might replace polycarbonate ones, but to test the ability to use quartz filters for INP analysis in stations where quartz filters are sampled routinely, allowing to extend the coverage of INP databases. This would also allow us to obtain both the chemical composition and the INP concentration from the same filter. We thank the reviewer for acknowledging the potential impact of the manuscript and we understand their concerns on the methods and the interpretations made. We have included several tests that support many statements made in the manuscript and that assure the reliability of the methods. We also identified the different aerosol conditions for every sampling day and stated the limitations of the evaluation of the three different methods for these particle types or specific aerosol conditions. All these general concerns have been considered when revising the manuscript and are more carefully addressed in the specific comments below as well as in the response to Reviewer #2.

A first conceptual issue is that the filter-method evaluation relies almost entirely on ambient aerosol samples. While ambient samples are valuable for method demonstration, a rigorous assessment of sampling and extraction methods should begin with controlled tests using laboratory-generated standard INPs (e.g., mineral dust, biological particles). The suitability of each method depends on INP type and the freezing temperature range, and this dependence cannot be separated using ambient mixtures alone. At minimum, the ambient dataset should span a wider range of atmospheric

conditions (e.g., clean days, dust events, heavy pollution, coastal influence). The manuscript should clearly acknowledge these limitations and specify the conditions under which the quartz-filter methods are applicable.

We agree with the reviewer on the fact that the methods should be evaluated in the laboratory, for different aerosol standards and under controlled conditions as we highlighted in the Conclusions section. As we also discuss in comment 41 of Reviewer #2, these laboratory evaluations were not performed at the AIDAd cloud chamber at the moment due to time limitations, but in light of the promising results obtained for ambient samples, we contemplate a potential campaign at this facility to further test the different sampling and particle extraction methodologies under controlled conditions.

Concerning the ambient samples, we performed a 3-month campaign where a total of 27 samples were collected under varying aerosol conditions (i.e., predominance of different aerosol types, higher/lower aerosol load,…). To understand the range of aerosol conditions during this measurement period, Figure R1 shows the time series of the equivalent black carbon mass concentration (eBC) measured at 880 nm with an aethalometer (AE33, Aerosol Magee Scientific), the integrated aerosol light-scattering coefficients at 450, 550 and 700 nm measured with a nephelometer (3563, TSI Inc.), the Scattering Angstrom Exponent (SAE) between 450 nm and 700 nm as well as the ratio of particle number concentration of fine and coarse particles (N_{Fine}/N_{Coarse}) measured with an Aerodynamic Particle Sizer (APS, 3321, TSI Inc.). While the eBC is an indicator of anthropogenic pollution in the station (mainly road traffic, Lyamani et al., 2011) and the scattering coefficient can be used as a proxy of aerosol load, the SAE indicates the predominant size of the aerosol population with values <1 indicating a predominance of coarse particles while values >1.5-2 indicate predominance of fine particles. This presence of fine or coarse mode particles can also be identified by the N_{Fine}/N_{Coarse} ratio.

We would like to highlight that we have only included data for the dates corresponding to filter sampling days, with the dots representing the 24h average and the error bars the standard deviation. We have indicated the aerosol conditions during each day as a colored shaded area around the markers.

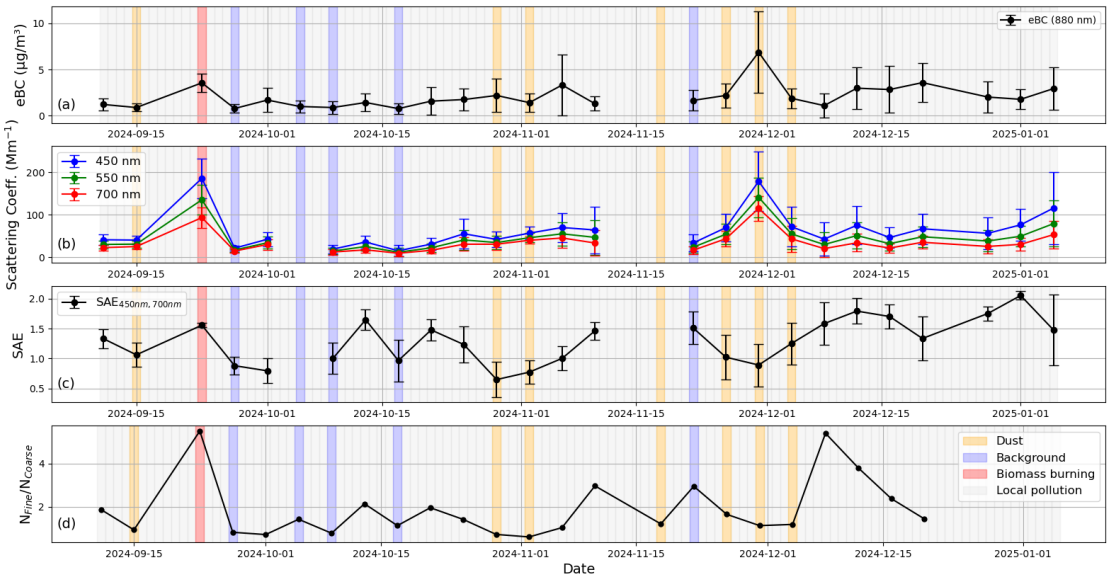

Figure R1. Time series of the daily averages of the eBC at 880 nm (a), the scattering coefficients at 450, 550 and 700 nm (b), the SAE between 450 and 700 nm (c) and the N_{Fine}/N_{Coarse} ratio (d). Error bars represent the standard deviation of the daily averages.

From Figure R1 we can identify three main predominant aerosol conditions during the period: local pollution, background conditions and dust events. Pollution days can be associated with moderate values of the scattering coefficient (≈ 50 Mm^{-1}) and large SAE values (≈ 1.5), where the eBC shows different values depending on the level of pollution. On the other hand, there are several days where the scattering coefficients present low values (≈ 20 Mm^{-1}) and low eBC (≈ 1 μgm^{-3}), which are associated with cleaner conditions in the station, which we have named background conditions. Also, given the proximity of the

city of Granada to the Sahara Desert, dust intrusions in the station during the year are very common and are characterized by high scattering, low SAE (generally < 1) and low N_{Fine}/N_{Coarse} ratios. From Figure R1 we can see that the majority of the filter sampling days correspond to pollution days, with five days of background conditions (27-09-2024, 05-10-2024, 09-10-2024, 17-10-2024, 22-11-2024). We can also identify several days where dust is dominant in the atmosphere, highlighting two major dust events, one of them happening on the 29-10-2024 and the other one covering the period from 26-11-2024 to 04-12-2024 with a strong peak on the 30-11-2024. Additionally, the time series shows one peak in scattering coefficients on 23-09-2024, which corresponds to a biomass burning event transported from the Western Iberian Peninsula coming from the wildfires in Portugal during September 2024, as confirmed with satellite and ancillary measurements at the station (not sown).

We have compared the performance of the different sampling methods (96-punch quartz, punch quartz, and polycarbonate) as a function of the aerosol conditions. The individual comparison of methods (shown in Figure S11 in the supplementary material) shows that in general there is good agreement between methods for most pollution days, with some differences during specific days where the INP concentration from the Polycarbonate method shows larger values at high temperatures. For background conditions, there is good agreement for the cases on 27-09-2024, 05-10-2024, 09-10-2024 and 17-10-2024, with some small discrepancies at high temperatures. However, the comparison on 22-11-2024 shows systematically lower INP concentrations for the Polycarbonate method than for the Quartz methods.

Concerning dust events, the comparison of the three methods shows different levels of agreement depending on the day. For the event on 29-10-2024 there is general good agreement between the three methods at high temperatures, but as the temperature decreases the Quartz punch washed method shows larger INP concentration compared to the other two methods. On the other hand, for the more intense dust event in late November, there is good agreement on the peak day (30-11-2024) especially at temperatures below-12 ºC. However, during the other two days (26-11-2024 and 04-12-2024) the behavior of the INP concentration of the Polycarbonate method is different from the one obtained with the Quartz methods, which might be due to the differences in the particles being sampled due to the PM10 cutoff for quartz filters. However, APS measurements from these days barely show particles over 10 μm in diameter, so these differences could then be related to the easiest extraction of these dust particles with polycarbonate filters. To confirm this, further laboratory experiments with these types of particles need to be performed.

Lastly, the biomass burning event shows good agreement in INP concentration for the three methods, although there are some small discrepancies between the Polycarbonate method and the Quartz method for temperatures over -12 ºC.

We would like to note that although the city of Granada is very close to the Mediterranean Sea, the orographic situation limits the influence of coastal particles (Titos et al., 2012; Titos et al., 2014), so the comparison presented in this work cannot be extrapolated to this type of aerosol particles.

To further evaluate the general validity of the comparison between methods with atmospheric samples, we have evaluated the differences in INP concentration between methods at three different temperatures for each atmospheric scenario described. Figure R2 shows the boxplots of the INP concentration difference at -10, -13 and -16 ºC, using the polycarbonate method as reference. We have selected these temperatures so that there is enough data to perform the statistical analysis (note that for biomass burning there is only one case). As expected from the results shown in the manuscript, the larger differences in INP concentration correspond to lower temperatures. However, from Figure R2 we observe that the differences in INP concentration at each temperature are very similar for every atmospheric scenario. This suggests that larger differences in INP concentration among methods are not related to a specific type of particle, at least for the aerosol conditions during this campaign. Still, as the reviewer has stated, further laboratory tests with aerosol standards are necessary to fully evaluate the INP concentration differences attributed only to the filter substrates and the extractions methods. So far, it seems that the results are not dependent on aerosol type and might be of general applicability under the specific aerosol conditions covered in our study (pollution/biomass burning, background conditions and dust events). However, since dust particles might differ from one event to another, we believe that further characterization of the methods with dust particles that are well characterized in size and chemical composition is necessary.

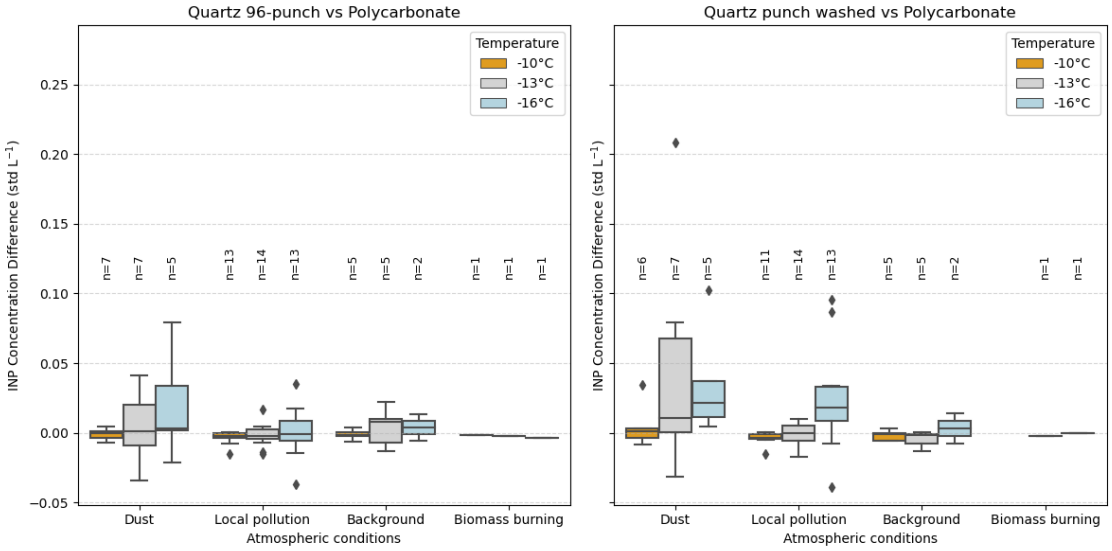

Figure R2. Boxplots of the INP concentration absolute differences between methods at three different temperatures (-10, -13, -16 ºC) for four different aerosol conditions. Horizontal lines represent the median values, lower and upper limits in the boxplots correspond to the first (Q1) and third (Q3) quartiles, where the difference between them represents the inter-quartile range (IQR). Lower and upper whiskers show Q1-1.5IQR and Q3+1.5IQR, respectively. Outliers are shown as diamond markers.

We have included this discussion in Section 4.3 as well as Figure R1 in the SI and stated the limitations of the results presented here in this section and in the conclusions.

L603-608: "Overall, the freezing spectra derived from the three methods agree at temperatures above -15 ºC, and measurements are mostly within standard error. In the range -10 to -7.5 ºC the INP concentrations derived from the Quartz methods are lower than those from the Polycarbonate method. When looking at the individual spectra in Figure S11 one can see that this feature is present during some specific days of the sampling period, where most of them correspond to pollution days or dust events."

L629-656: "The individual comparison of the INP concentrations (shown in Figure S11) involved in the average from Figure 6 shows that the agreement between methods differs from day to day, which could originate from the nature of the particles in the sample. To assess this, we identified the dominant particle type on each sampling day based on aerosol optical and microphysical properties measured at the same station. We have analyzed the equivalent black carbon concentration (eBC), the ratio of concentration of fine and coarse particles, the scattering coefficients and the scattering Ångström exponent (SAE) at the surface, which are shown in Figure S12. The analysis of the optical properties revealed that the majority of the sampling days corresponded to pollution days, with a few cases of cleaner/background conditions at the station. Additionally, there are several dust events and a biomass burning event (transported from Portugal) happening in the measurement period. Even though the city of Granada is near the Mediterranean Sea, the orographic situation limits the contribution of marine aerosol to the aerosol population (Titos et al., 2012; Titos et al., 2014), so the results shown are only representative of these aerosol conditions. We found that there is a general good agreement for most pollution events, with some differences during specific days where the INP concentration from the Polycarbonate method shows larger values at high temperatures. The biomass burning event shows a very good comparison of the three methods, as happened in most of the pollution days. For background conditions, there is also good agreement between methods apart from one day where the Polycarbonate method shows systematically lower INP concentrations. Lastly, concerning dust events, the comparison of the three methods shows different levels of agreement depending on the day. When the dust concentration is very high, we find that there is some disagreement between the methods at high temperatures, with higher INP concentrations for the Polycarbonate method. Even though this might be related to the differences in size cutoff in the sampling line, which can be especially relevant for dust, we did not observe a large contribution of particles larger than 10 μm in diameter in the size distribution measurements, so these differences might be related to a more efficient extraction of the dust particles with polycarbonate filters compared to quartz filters. Overall, results presented do not seem dependent on aerosol

type and might be of general applicability under the specific aerosol conditions covered in this study (pollution/biomass burning, background conditions and dust events). However, since dust particles might differ from one event to another, further characterization of the methods with dust particles that are well characterized in size and chemical composition is necessary."

A second major concern relates to the extraction methodology. For a methods paper, key parameters such as droplet volume, extracted filter area, extraction time, and extraction technique require thorough justification. Many immersion-freezing instruments, including CSU-IS and INSEKT, use 50 µL droplets. The authors use 100 µL droplets, but the implications of this choice are not sufficiently discussed. Likewise, the manuscript states that particles were extracted using "manual agitation" for 60 seconds, but the procedure is not described in sufficient detail to evaluate its reproducibility or effectiveness. Such a short extraction time and uncontrollable extraction method is particularly problematic for quartz fiber filters, whose porous structure retains particles much more strongly than smooth PC filters. A clearer description, supporting evidence, and discussion of potential biases are needed.

We understand the reviewer's concerns about the extraction methodology. Regarding GRAINS' droplet volume, the use of a larger droplet volume causes a lower limit of detection (LOD) of the instrument, which has implications in the higher probability of detecting rare INPs and of course in the calculated INP concentration values itself, as well as their uncertainties. While it is true that GRAINS has a different droplet volume than many immersion-freezing instruments, there are other parameters that have the same implications, such as sampled volume of filters, which differ from one study to another, also contributing to the absence of a standardized protocol for determining the INP concentration with immersion-freezing techniques. Nevertheless, we have added information about the implications of GRAINS' droplet volume in the text, which is stated between L131-134 in the revised manuscript.

Concerning the manual agitation, we agree with the reviewer about the fact that there are more reproducible methods for extracting particles from the filters. At the time of these experiments, a rotating agitator was not available at our laboratory, which is the particle extraction method recommended by the ACTRIS Center for Cloud Ice Nucleation (CCIce) when using polycarbonate filters. Very recently our laboratory was equipped with this instrument and we performed several tests after the analysis of the samples to ensure that the manual agitation method used in our previous analysis is equivalent and reproducible and that we can rely on the measurements included in the manuscript.

In particular, we evaluated the two extraction methods for the polycarbonate filters, one being the manual agitation for 60 seconds and the other one being rotating the sample for 20 minutes at 60 r.p.m. in a rotating agitator (first we always manually agitate briefly to fully immerse the filter in water). To this end, we used three sets of two filters sampled simultaneously and analyzed following the two extraction methods described above. Figure R3 shows the comparison of these two extractions methods for three different sets of filters (each set of two filters sampled in the same day, under pollution conditions), where one can observe that the differences between the two methods are within uncertainties, so the manual agitation can be considered valid.

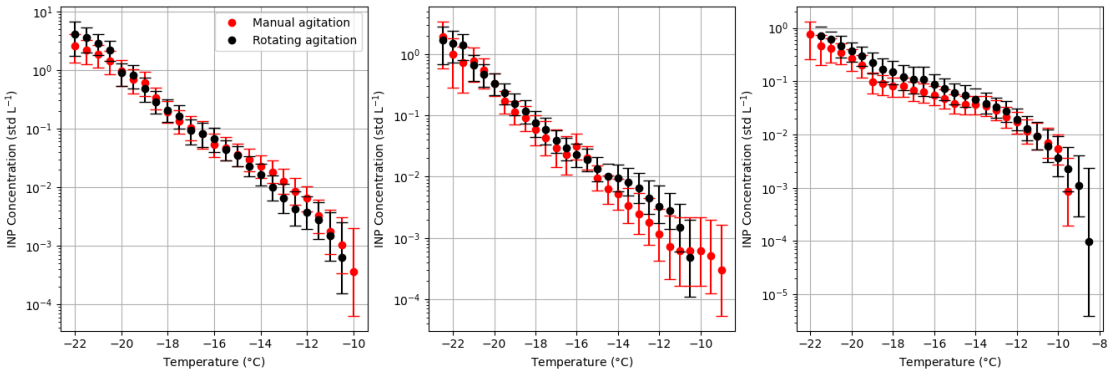

Figure R3. Comparison of the calculated INP spectra during different days depending on the particle extraction method for polycarbonate filters. Uncertainties have been calculated according to Agresti and Coull (1998).

Concerning the Quartz punch washed method, we first performed 5 experiments to determine the INP concentration with GRAINS using 1 cm punches from different regions of the same filter in order to test the variability of the method itself. Figure R4 shows the obtained INP concentrations, which are mostly within the uncertainties calculated following Agresti and Coull (1998) (right panel in Figure R4 shows the same data but without uncertainties for clarity). Once we acquired the rotating agitator, we did the same tests as with the polycarbonate filters to compare both extraction methods, manual and rotating agitation. We used three filter samples collected on different days (under dust and background conditions) and did two 1 cm punches in each filter. The results are shown in Figure R5. Again, one can observe that both extraction methods lead to similar INP concentrations and are within uncertainties.

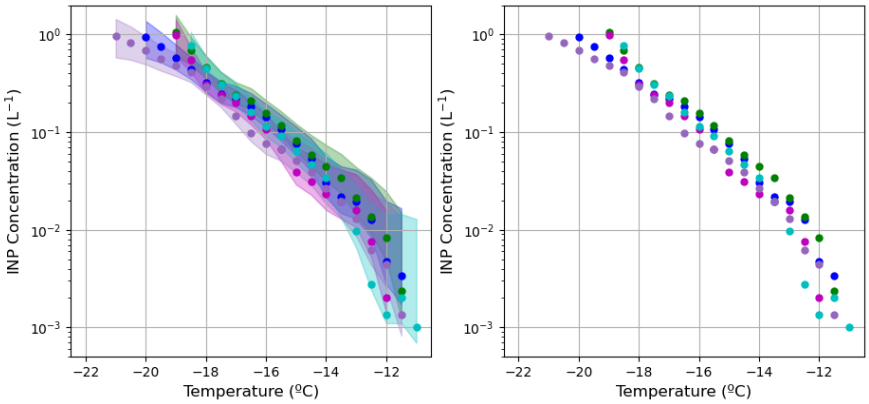

Figure R4. INP spectra obtained following the Quartz punch washed method for different regions of one quartz filter with uncertainties by Agresti and Coull (1998) (left panel) and without them (right panel).

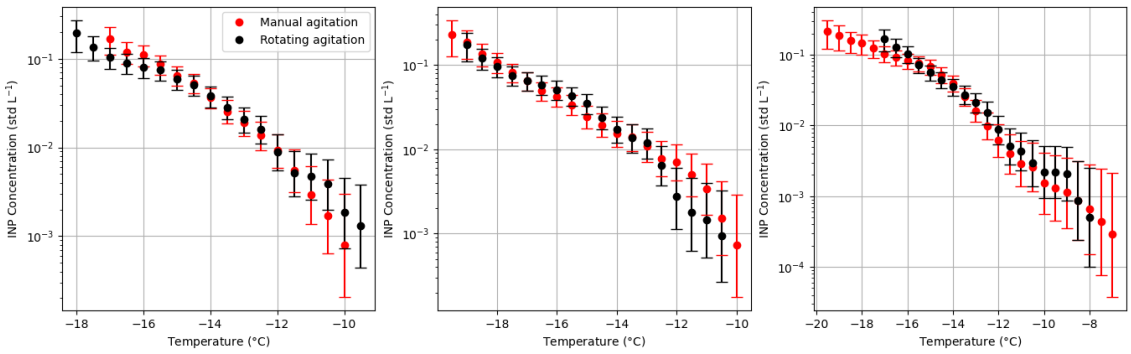

Figure R5. Comparison of the calculated INP spectra during different days depending on the particle extraction method for quartz filters (Quartz punch washed method). Uncertainties have been calculated according to Agresti and Coull (1998).

We are aware that differences in extraction efficiency might depend on the aerosol size and composition, and also on filter substrate as suggested by the reviewer. However, although limited, our intercomparison of extraction methods allowed us to be confident on the results shown in this manuscript concerning the manual agitation for both polycarbonate and quartz filters. Furthermore, we would like to highlight that at the moment, we perform the rotating agitation method for both the Polycarbonate and the Quartz punch washed methods, as it is recommended by CCIce and we agree with the reviewer in the fact that it is a more reproducible method for the extraction of particles from filters.

We have included this information in the methodology section, between L379-397 in the revised manuscript.

"After sample collection, polycarbonate filters were analyzed in the laboratory as explained in Section 3.1 (Polycarbonate method). The extraction of the particles was done by manually agitating the tube containing the filter immersed in 20 mL of filtered ultrapure water for 1 min. The recommended protocol established by the ACTRIS Center for Ice Nucleation (CCIce) for particle extraction consists in rotating the tube with the suspension in a rotating agitator for 20 min at 60 r.p.m (CCIce 4th QAQC workshop). To ensure the equivalence of the INP concentrations obtained by manual and rotating agitation, several tests were performed, leading to very similar results. For quartz filters, we followed two approaches. For the first approach (Quartz 96-punch), we punched a region of the filter 96 times with the use of a biopsy punch of 1 mm diameter, we introduced each punched region into the wells of the PCR and then we filled them with 100 µL of filtered ultrapure water. This approach is chosen in many INP studies (Tatzelt et al., 2022; Welti et al., 2018; Wex et al., 2019), since it only requires a small fraction of the quartz filter and the rest of it can be used for additional analyses. The second approach (Quartz punch washed) consisted of punching a portion of the filter with a 1 cm diameter biopsy punch and then washing it in filtered ultrapure water in a similar way as typically done with polycarbonate filters, by manually agitating the tube. This approach was followed in Bras et al., (2024) and Lacher et al. (2024) for obtaining INP concentrations with the LINDA instrument (Stopelli et al., 2014). Again, we performed several tests to verify the similarity between manual and rotating agitation and to assess the reproducibility of manual agitation, obtaining equivalent results. To obtain the INP concentration with the Quartz 96-punch method we have used Equation 6, whereas for the Polycarbonate and Quartz punch washed methods we calculated the INP concentration using Equation 3."

Related to this, extraction efficiency is a central consideration in comparing sampling substrates. PC filters are widely used because they allow collected particles to be washed off efficiently. Quartz filters, in contrast, consist of a fibrous matrix in which particles can become embedded, resulting in lower extraction efficiency. The manuscript should discuss how this intrinsic structural difference may influence INP recovery, especially for the punch-washed method. In that method, lots of quartz fibers are inevitably transferred into the suspension and can themselves act as INPs at lower temperatures (Conen et al. 2012; Harrison et al., 2019). Their contribution must be carefully considered.

The reviewer is right, when applying the Quartz punch washed method there are fibers that are transferred to the suspension, as happens with the individual punches in the Quartz 96-punch method that are introduced in the wells and then filled with ultrapure water. Concerning the particle extraction efficiency of the Quartz punch washed method, we would like to highlight that the section of the filter that we wash in water 'falls apart' and does not stay intact as it happens with the polycarbonate filters.

As for the possible contribution of these quartz fibers to the INP concentration, we have evaluated the ice nucleation ability of the blank filters of each method. The results can be observed in Figure R6 (Figure S5 in the supplementary material). We would like to highlight the resemblance between the frozen fraction values at lower temperatures of both quartz methods with that of water, implying that the quartz fibers do not contribute greatly to the INP concentration at lower temperatures. In fact, the blank filters for the Quartz punch methods show more similarity to the water curve than the blank filters for the Polycarbonate method, suggesting that the contribution of INPs is higher for the latter. However, regardless of the method used, background signals of the filters should be routinely examined to guarantee the quality of the measurements.

Also, looking at the individual comparison of methods in Figure S11, one can see that there are many cases where the INP concentration obtained with the three methods is identical, and the Quartz punch washed method tends to show the higher INP concentration values (which from Figure S5 we consider that do not come from the quartz fibers), so we believe that the particle extraction in this method is efficient.

We have included this discussion between L688-698 in the manuscript:

"This was expected by looking at Figure 6, where the average INP concentrations obtained with the quartz samples were higher than those obtained with the polycarbonate samples. According to Conen et al. (2012), the fibers from quartz filters do not contribute to the INP concentration at temperatures higher than -12 ºC. On the other hand, from Harrison et al. (2019) one could expect that the quartz fibers could act as INP at lower temperatures, and this could explain the differences observed in our results for the Quartz methods

as compared to the Polycarbonate method. However, as observed in Figure S5, the average frozen fraction curves of the handling filters (6 experiments per method) show lower values for both Quartz methods, so in our case it seems that there is very little contribution of the quartz fibers to the total INP concentration, as extracted from Figures 6 and S11. Still, the background signals of the filters should always be analyzed to guarantee the accuracy of the INP measurements."

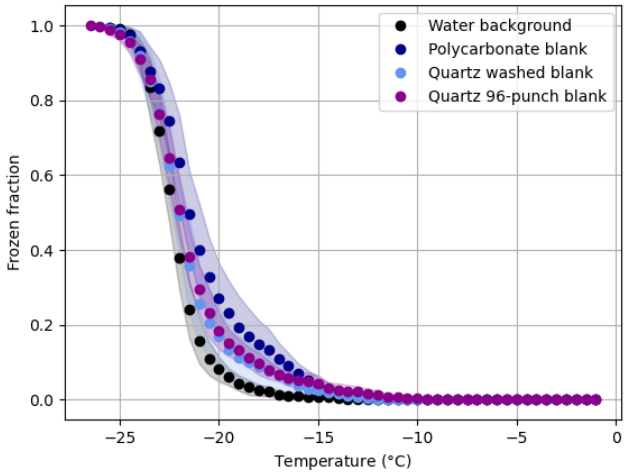

Figure R6. Frozen fraction of the different filter backgrounds for each method. Data shown correspond to averages from 6 different experiments, where shaded area corresponds to the standard deviation.

These methodological issues directly affect the interpretation of the key results. For example, below approximately -12 °C, the manuscript reports higher INP concentrations for the two quartz-based methods than for the PC method. The explanation offered in the manuscript remains speculative and lacks supporting evidence. A more plausible interpretation, consistent with Conen et al. (2012) and subsequent studies, is that the quartz filters introduce additional quartz fibers that act as INPs at colder temperatures. This naturally leads to increasing discrepancies among methods as temperature decreases. Indeed, Conen et al. (2012) concluded that quartz punch method is a better choice and should be restricted to temperatures ≥ -12 °C. This important limitation should be explicitly discussed. In this context, it is also worth noting that the quartz 96-punch method is likely the cleanest configuration, with minimal fiber contamination; the very short 60-second agitation step may, however, lead to insufficient particle extraction. This distinction is important because the 96-punch method is the one recommended by Conen et al. (2012) and Wex et al. (2019). Therefore, statements in the manuscript suggesting that the punch-washed method may be preferable are potentially misleading and require substantial reconsideration or additional solid experimental evidence.

Again, we would like to refer to the results obtained for our blank filters. If we apply subtraction of the background signal (instead of water subtraction of ultrapure water) the calculated INP concentrations are slightly lower for all methods. However, the differences among methods remain basically the same due to the similarity of the background signals.

Also, we would like to highlight that the amount of surface of quartz filter involved in both Quartz punch methods is of the same order, with 0.75 cm^2 for the Quartz 96-punch method and 0.79 cm^2 for the Quartz punch washed method. Therefore, we can assume that the amount of fiber involved in both methods is practically identical. Furthermore, we have found that the blank filter for the Quartz 96-punch method presents an average frozen fraction with slightly larger values than the one for the Quartz punch washed method, so in principle the cleanliness of the two Quartz punch methods is equivalent.

As for the particle extraction discussion, we refer the reviewer to the second comment.

In summary, while the instrument development and the general methodological framework are strong, several core conclusions regarding the equivalence and relative performance of the filter methods are currently not yet supported by the available data. I encourage the authors to

substantially revise the manuscript, explicitly state the limitations, incorporate a more cautious interpretation of the results, and restrict conclusions to the conditions actually tested.

As the reviewer suggested, we have included the limitations of the conditions tested in the manuscript, stating that laboratory experiments with different types of particles are needed to further evaluate the different methods. We have rewritten parts of the conclusions including all the suggestions made by the reviewer that have been discussed in the previous comments and softened some of our take-home messages.

Specific comments:

1. **L42: "ice-nucleating particles (INPs)"**

   Thanks, we have corrected this.

References:

Conen, F.; Henne, S.; Morris, C. E.; Alewell, C., Atmospheric ice nucleators active ≥ −12 °C can be quantified on PM10 filters. Atmospheric Measurement Techniques 2012, 5, (2), 321-327.

Harrison, A. D.; Lever, K.; Sanchez-Marroquin, A.; Holden, M. A.; Whale, T. F.; Tarn, M. D.; McQuaid, J. B.; Murray, B. J., The ice-nucleating ability of quartz immersed in water and its atmospheric importance compared to K-feldspar. Atmospheric Chemistry and Physics 2019, 19, (17), 11343-11361.

Wex, H.; Huang, L.; Zhang, W.; Hung, H.; Traversi, R.; Becagli, S.; Sheesley, R. J.; Moffett, C. E.; Barrett, T. E.; Bossi, R.; Skov, H.; Hünerbein, A.; Lubitz, J.; Löffler, M.; Linke, O.; Hartmann, M.; Herenz, P.; Stratmann, F., Annual variability of ice-nucleating particle concentrations at different Arctic locations. Atmospheric Chemistry and Physics 2019, 19, (7), 5293-5311.

Lyamani, H., Olmo, F. J., Foyo, I. and Alados-Arboledas, L. (2011). Black carbon aerosols over an urban area in south-eastern Spain: Changes detected after the 2008 economic crisis, *Atmos. Environ*., 45(35), 6423-6432, https://doi.org/10.1016/j.atmosenv.2011.07.063

Titos, G., Foyo-Moreno, I., Lyamani, H., Querol, X., Alastuey, A., and Alados-Arboledas, L. (2012), Optical properties and chemical composition of aerosol particles at an urban location: An estimation of the aerosol mass scattering and absorption efficiencies, *J. Geophys. Res*., 117, D04206, https://doi.org/10.1029/2011JD016671

Titos, G., Lyamani, H., Pandolfi, M., Alastuey, A., & Arboledas, L. (2014). Identification of fine (PM_1) and coarse (PM_{10-1}) sources of particulate matter in an urban environment. *Atmos. Environ.*, *89*, 593–602. https://doi.org/10.1016/j.atmosenv.2014.03.001