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.

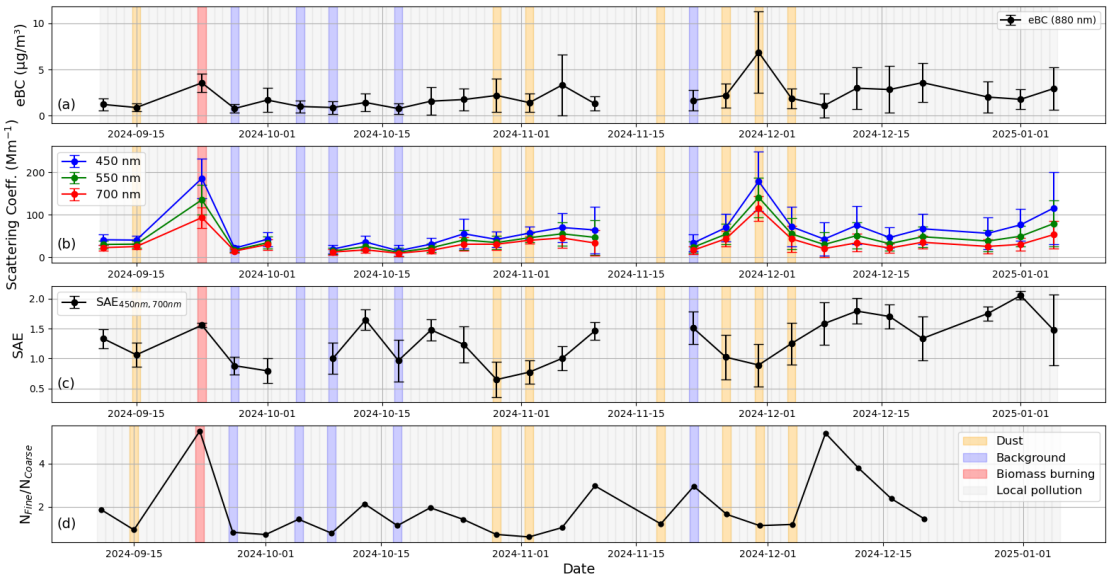

Figure R1. Time series of the daily averages of the eBC at 880 nm (a), the scattering coefficients at 450, 550 and 700 nm (b), the SAE between 450 and 700 nm (c) and the N_{Fine}/N_{Coarse} ratio (d). Error bars represent the standard deviation of the daily averages.

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.

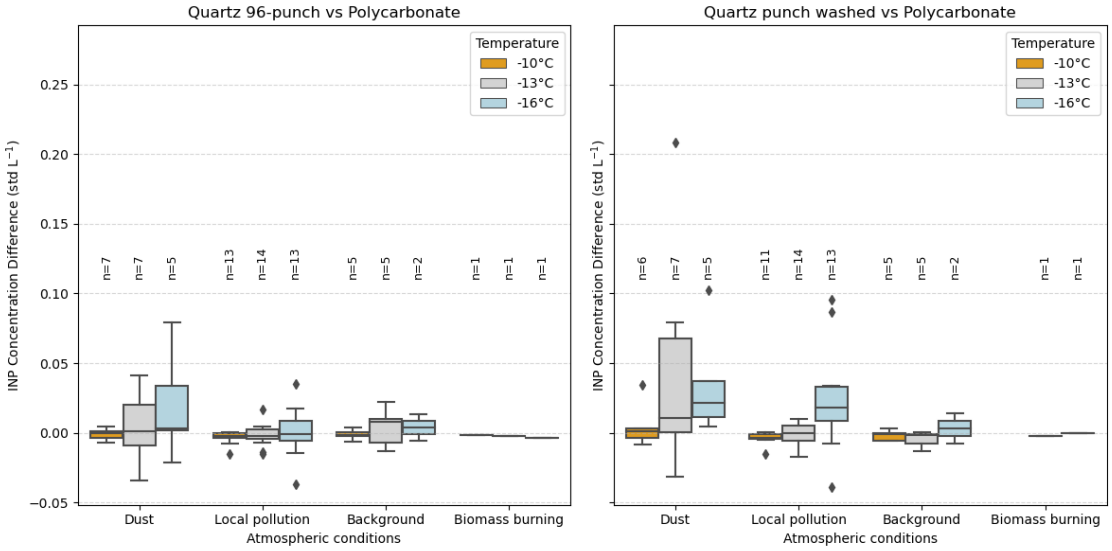

Figure R2. Boxplots of the INP concentration absolute differences between methods at three different temperatures (-10, -13, -16 ºC) for four different aerosol conditions. Horizontal lines represent the median values, lower and upper limits in the boxplots correspond to the first (Q1) and third (Q3) 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.

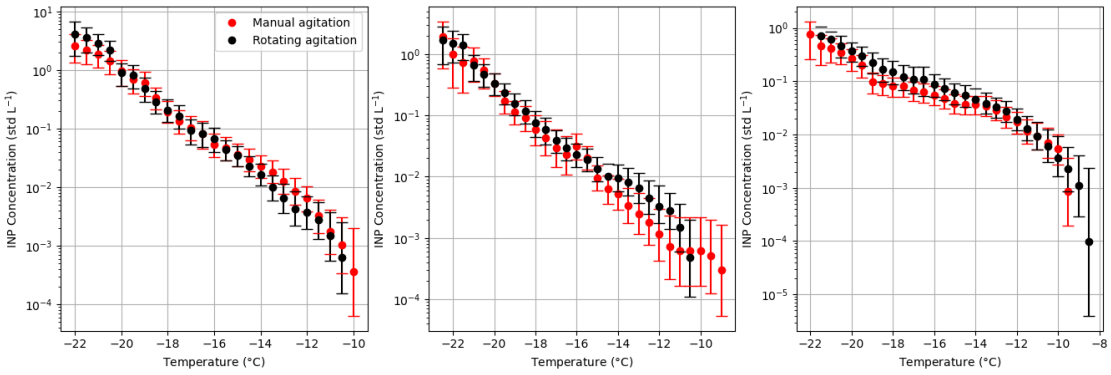

Figure R3. Comparison of the calculated INP spectra during different days depending on the particle extraction method for polycarbonate filters. Uncertainties have been calculated according to Agresti and Coull (1998).

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.

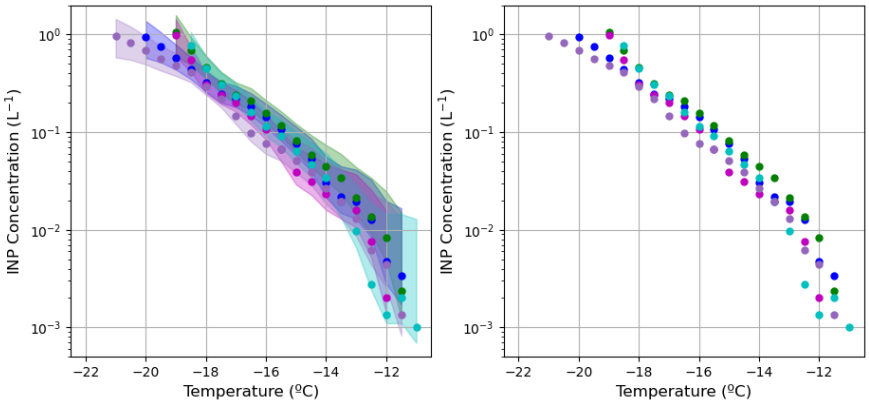

Figure R4. INP spectra obtained following the Quartz punch washed method for different regions of one quartz filter with uncertainties by Agresti and Coull (1998) (left panel) and without them (right panel).

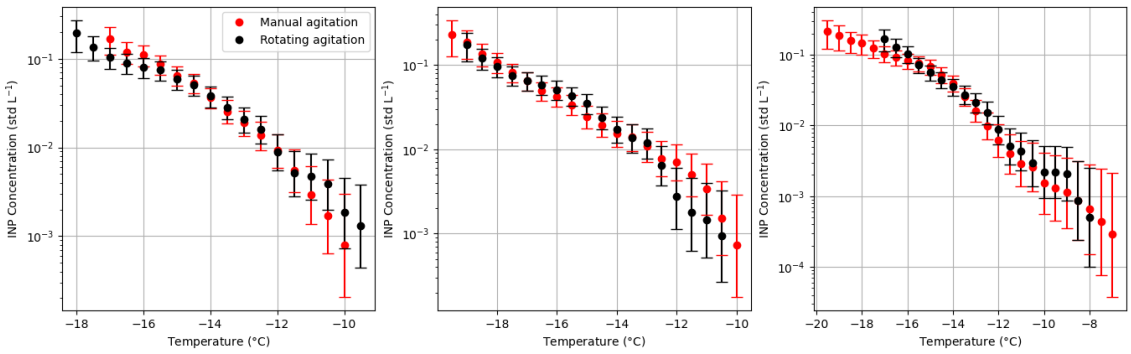

Figure R5. Comparison of the calculated INP spectra during different days depending on the particle extraction method for quartz filters (Quartz punch washed method). Uncertainties have been calculated according to Agresti and Coull (1998).

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."

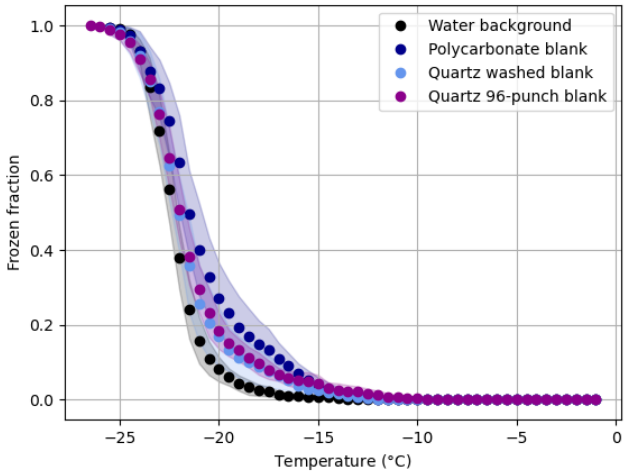

Figure R6. Frozen fraction of the different filter backgrounds for each method. Data shown correspond to averages from 6 different experiments, where shaded area corresponds to the standard deviation.

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

Response to Reviewers

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Reviewer #2

Bazo et al. demonstrate and validate a new immersion freezing analysis platform for the measurement of INPs, the GRAINS, based on the CSU-Ice Spectrometer. The authors performed a comprehensive set of tests against other INP instruments to confirm its comparability and accuracy. Some of these tests also highlighted discrepancies between some instruments for some materials, such as between PINE and immersion freezing instruments for K-feldspar. Importantly, the authors demonstrated that the use of three filter sampling techniques, including traditional polycarbonate washing and quartz punch techniques, yield similar results, although there were some discrepancies at colder temperatures. This work highlights the need to compare different sample collection techniques to ensure consistency in INP measurements. These findings warrant publication in AMT pending the following revisions and comments.

Major comment:

Section 4.1: Often, new instruments are validated using a range of lab prepared samples that cover a wide temperature, e.g. Snomax, NX illite, K-feldspar. I would have expected to see a few examples, particularly given the discrepancies that can be encountered with NX illite, while acknowledging as the authors state that there are no reference standards for immersion freezing and the materials that the community uses all have some variability. This is perhaps more surprising given that multiple other samples (including K-feldspar and ATD) are tested as part of intercomparisons – in principle it should be possible to compare K-feldspar and ATD to literature data, although it may not be quite a 1-to-1 comparison since these were aerosolised rather than being prepared as suspensions directly from lab samples.

The reviewer is completely right about the use of different samples to validate new freezing instruments. We used NX Illite for validation and, indeed, we considered the use of additional samples, such as Snomax, to validate GRAINS at a higher temperature range. However, since this sample is biological, we realized that it can be easily degraded and therefore have different ice nucleation properties as reported by studies such as Wex et al. (2015). This is why we considered that performing direct intercomparisons with other INP devices would be a better approach to validate GRAINS, with both ambient particles and reference samples.

Nevertheless, we agree with the reviewer on the fact that the freezing ability of K-feldspar and ATD as presented in the manuscript could be also compared to literature data. Figure R1 shows results of the ice nucleation properties of the four samples compared to existing parametrizations of K-feldspar, ATD and mineral dust, where we obtained the ice nucleation active sites (INAS) density (n_s) per unit of aerosol surface from AIDAd, PINE, INSEKT and GRAINS. In general, for ATD, SDSA01 and Sahara dust we observe a good agreement for all devices (this was expected for most of them given Figure 5 in the manuscript). The slopes of the n_s spectra follow the same trend as parametrization, but the experimental data shows slightly lower values of n_s.

For K-feldspar, we observe that the slope of the n_s obtained with GRAINS follows the parametrization by Atkinson et al. (2013), but the differences in the n_s values are around three orders of magnitude. The other experimental data also shows differences with respect to the parametrization by Atkinson et al. (2013), with AIDAd data being below the line and PINE data falling near the region of the A13 parametrization but with a slightly different slope. For INSEKT, the differences are very large when compared to literature data, but still, there is very good agreement with the experimental data by GRAINS. Our hypothesis is that the droplet freezing arrays might be underestimating the INP concentration of such a highly active substance because of the assumption of a single INP per droplet (Vali, 1971). Another option could be that the ice nucleation activity of K-feldspar is degraded due to being immersed in water. This is something observed by Harrison et al. (2016), although the degradation occurred after the sample was immersed in water for months. However, Kumar et al. (2018) stated that when K-feldspar is immersed in water there are interactions between the water and the sample's surface, which could explain the difference between the ice nucleation ability obtained with droplet freezing arrays (where K-feldspar is immersed in water for hours) and the one obtained with the expansion chambers. In any case, since the n_s spectra obtained with INSEKT and GRAINS is practically identical, and furthermore the comparison of both instruments with PINE and AIDAd is good for the rest of the samples, we trust that the differences for K-feldspar are caused by the nature of the sample rather than by any issue with our droplet freezing array. In these cases, we believe that for the validation of newly developed instruments, the intercomparisons between instruments would provide more information than a comparison with literature data.

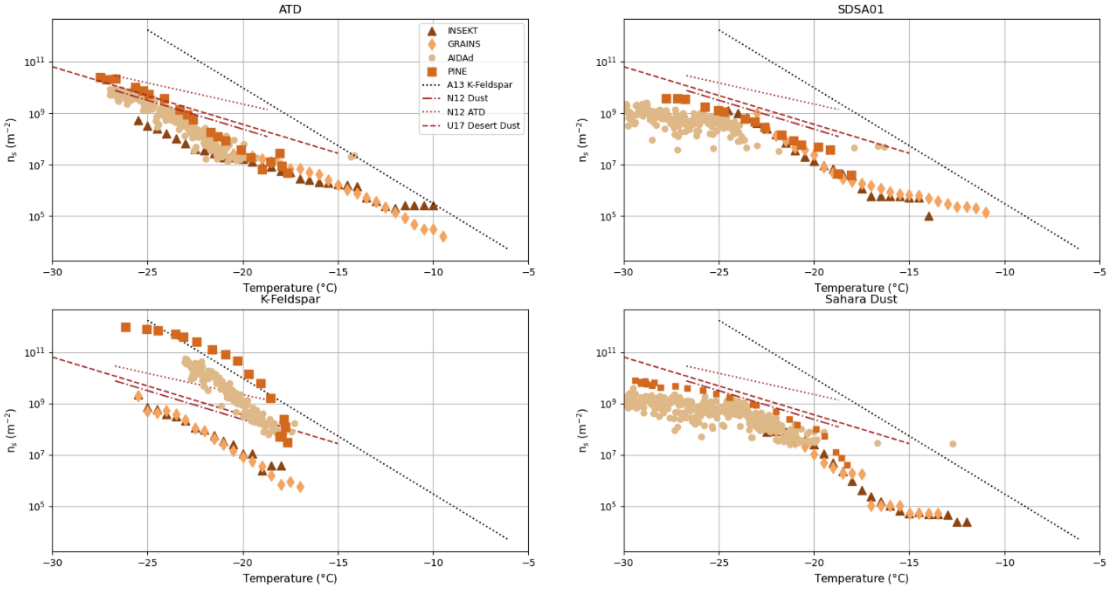

Figure R1. Ice nucleation active sites (INAS) density (n_s) per unit of aerosol surface obtained with INSEKT, GRAINS, PINE and AIDAd for ATD, SDSA01, K-Feldspar and Sahara Dust. Parametrizations from Atkinson et al. (2013) (A13), Niemand et al. (2012) (N12) and Ulrich et al. (2017) (U17) are also shown.

Concerning the data shown in the manuscript, we have changed Figure S10 in the SI, and now it shows the n_s by PINE, INSEKT and GRAINS as well as the parametrizations from the literature (Figure R6 in this document). We have also modified the text in Section 4.2 to include this discussion and added the new references to the reference list.

L542-546: "For the calculation of the total surface area necessary to obtain n_s from INP data, we applied a lognormal fit of the size distribution, as done in similar studies (e. g. Hiranuma et al. 2015), which was obtained with a combination of measurements from the Scanning Mobility Particle Sizer (SMPS) and the APS that operated at AIDAd."

L565-585: "Figure S10 also shows parametrizations reported in the literature for ATD (Niemand et al. 2012 – N12), K-Feldspar (Atkinson et al. 2013 – A13) and dust (Niemand et al. 2012 – N12; Ulrich et al. 2017 – U17). In general, the slopes of the n_s spectra for ATD, SDSA01 and Saharan dust follow the same trend as the dust parametrizations, but the experimental data shows slightly lower values of n_s, especially for both droplet freezing arrays. For K-Feldspar the two spectra overlap for the majority of the temperature range,

but PINE n_s values are larger than the ones obtained with the two droplet freezing arrays, by more than two orders of magnitude. In fact, PINE data shows n_s values similar to those corresponding to the A13 parametrization, whereas INSEKT and GRAINS show the same slope of the spectra as the parametrization by Atkinson et al. (2013), but with a difference of two order of magnitude in the values. This could be due to the fact that the working principle of INSEKT and GRAINS assumes that there is a single INP per droplet in the PCR wells (Vali, 2019), and since K-Feldspar is a highly ice-active substance the n_s obtained with both droplet freezing arrays could be underestimated. Another possibility could be that the ice nucleation activity of K-Feldspar is degraded due to being immersed in water. This is something observed by Harrison et al. (2016), although the degradation occurred after the sample was immersed in water for months. Also, Kumar et al. (2018) stated that when K-Feldspar is immersed in water there are interactions between the water and the sample's surface, which could explain the difference between the ice nucleation ability obtained with droplet freezing arrays compared to PINE and the A13 parametrization. Apart from the case of K-Feldspar, the results shown in Figure 5 and Figure S10 confirm the similarity of the two droplet freezing arrays and the PINE instrument, confirming that GRAINS can reproduce the freezing behavior of mineral dust samples when compared to INSEKT and PINE."

Minor comments:

1. **Line 28: Were the randomly punched quartz filters also washed?**

   They were not, the punched regions of the quartz filters were directly introduced into the wells of the PCR trays of GRAINS, as explained in the Methodology section.

2. **Line 35: Any indication of groupings? Were minerals more affected than biogenic materials (if analyzed) or vice versa?**

   This is a very interesting point that was also commented by Reviewer 1.

   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.

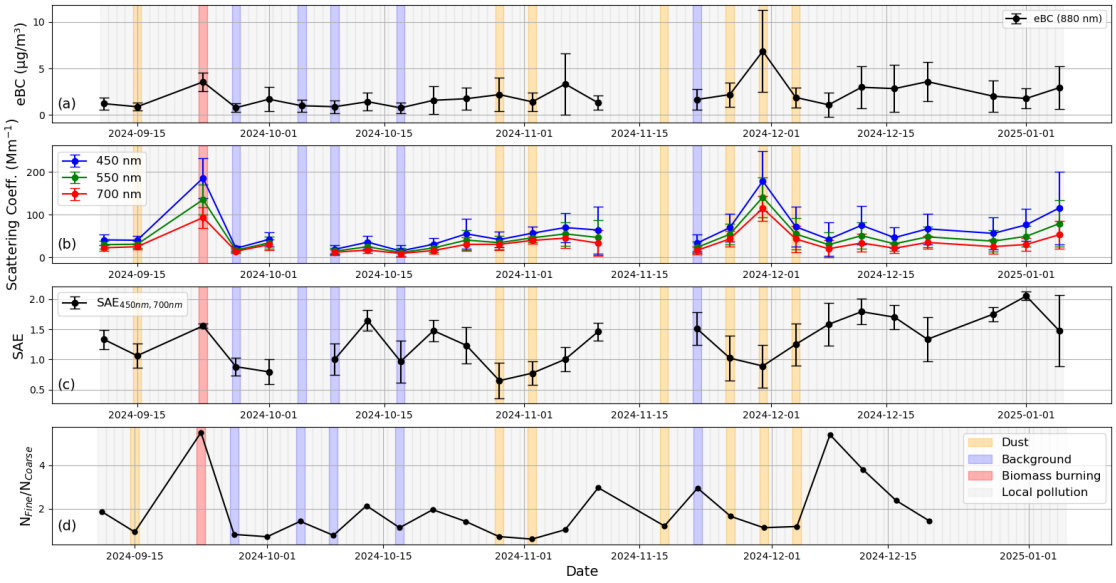

Figure R2. Time series of the daily averages of the eBC at 880 nm (a), the scattering coefficients at 450, 550 and 700 nm (b), the SAE between 450 and 700 nm (c) and the N_{Fine}/N_{Coarse} ratio (d). Error bars represent the standard deviation of the daily averages.

From Figure R2 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 R2 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 R3 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 R3 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.

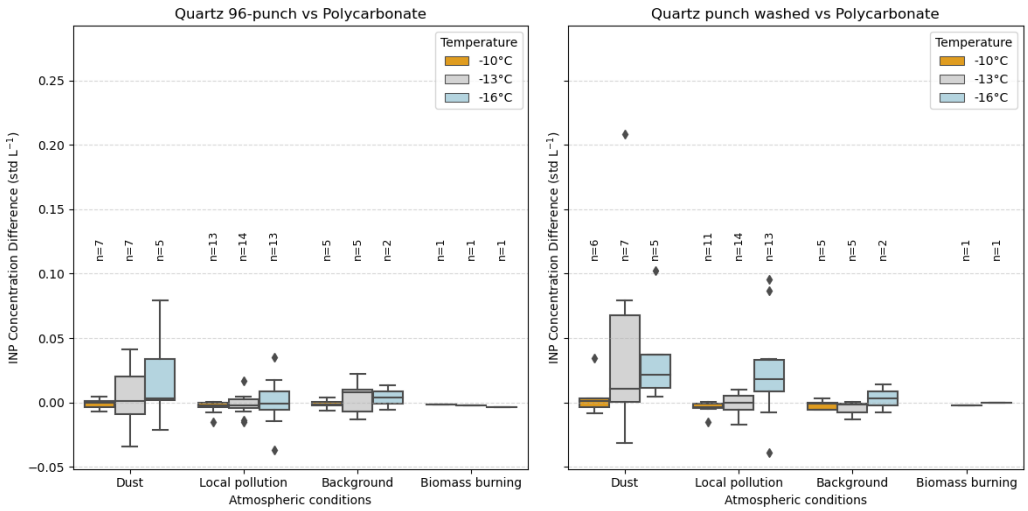

Figure R3. Boxplots of the INP concentration absolute differences between methods at three different temperatures (-10, -13, -16 ºC) for four different aerosol conditions. Horizontal lines represent the median values, lower and upper limits in the boxplots correspond to the first (Q1) and third (Q3) 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 R2 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."

3. **Line 37: Is the quartz 96 punch method a standard method?**

The Quartz 96 punch method is widely used in the scientific community (e. g. Tatzelt et al., 2022; Welti et al., 2018; Wex et al., 2019), as an alternative to the Polycarbonate method.

4. **Line 42: If "ice-nucleating particles" are being discussed in plural, then the abbreviation should be "INPs".**

Thanks for the note, we have changed it.

5. **Lines 56-59: CFDCs and PINE are mentioned (I would recommend explicitly mentioning PINE here, particularly since it is used in the study), alongside "INP spectrometers" – a very brief description of these modes of operation should be provided here, particularly of the "spectrometers" and especially since this is not necessarily a term that is used consistently throughout the community.**

We thank the reviewer for this suggestion. We have changed the term 'INP spectrometer' to 'droplet freezing array' throughout the text, a more common term used in the scientific community. We have also modified this part of the text as (L58-64):

"or cloud expansion chambers like the Portable Ice Nucleation Experiment (PINE; Möhler et al., 2021), in which the INP concentration can be measured in real time with a high temporal resolution. However, the majority of instruments to study INPs are offline techniques, mainly due to the simpler and less expensive instrumentation involved. In particular, one of these offline techniques are droplet freezing arrays, which cool down droplets of an aqueous solution containing INPs until they freeze (Creamean et al., 2025; David et al., 2019; Ladino et al., 2022; Wang et al., 2025; Wieber et al., 2024)."

6. **Line 61: They are also require far simpler and much cheaper instrumentation, and often easier to use and easier data to process.**

We have added this information in L60.

7. **Line 64: As in the above comment, there has been no description of how the technique operates in general (i.e. cooling droplets of aqueous suspensions of INPs on a cold stage until they freeze) so this discussion of different droplet volumes does not make much sense.**

As we modified the text in comment 5 explaining more about the technique of droplet freezing arrays, we believe that this discussion should remain in the text.

8. **Line 77: For the layperson, it might be confusing that there is not consistency across the instruments being used, e.g. why doesn't everyone use the same instrument, or why are new but similar instruments still being developed? Within the community, we know that this is because no such "standard" cold stage instrument exists, and so every group must build their own and test it. It would be worth noting this here.**

The reviewer is right; we have indicated that these types of instruments are typically developed by each research team (L81).

9. **Line 86: Are INP spectrometers specifically defined as being those that use PCR plates? As opposed to other cold stage techniques?**

There are several INP devices, such as CSU-IS or INSEKT, which are defined as INP spectrometers and do use PCR plates in their setup. However, there is no concise definition of this term, so we have replaced the term 'INP spectrometer' to 'droplet freezing arrays' throughout the text, as already stated in comment 5.

10. **Line 93: Replace "the first one" with "the polycarbonate filter washing method" or similar.**

Done.

11. **Line 96: What is meant by "a bigger punch"? What size is the punch?**

We wanted to highlight that in the mentioned studies (Bras et al., 2024; Lacher et al., 2024), where a punch of the quartz filter is washed, this punch is bigger than the ones used in Conen et al., (2012) and Wex et al., (2019). We have indicated the size of the punch used in Bras et al., (2024) and Lacher et al., (2024), which is 1.2 cm in diameter.

12. **Figure 1 caption: Some more description is needed. Why are some of the wells red in the image?**

These wells are the ones used in the experiments to characterize GRAINS' temperature. We have added this information to the figure caption (L128-129):

"Figure 1. Schematic of the GRAINS instrument. Red wells correspond to the ones used for temperature characterization described in Section 2.3."

13. **Line 109: The citation for Perez Fogwill 2024 is not in the reference list and I cannot immediately find it online.**

We apologize for the mistake, the reference corresponds to a poster contribution at the European Aerosol Conference 2024 in Tampere (Finland). The abstract can be found in page 788 of the Conference's abstract book

(https://www.dropbox.com/scl/fi/mk12y4q0ux3rnqrjo1nqj/EAC2024_Abstract_Book.pdf?rlkey=19grqfz3ec8lsuyvcu83e6iun&e=2&st=soar36hh&dl=0). We have added this reference to the reference list (L1007-1009):

"Perez Fogwill, G., Nontasin, P., Piedehierro, A. A., Mustonen, L., Welti, A. (2024). The Sample Volume Effect on Observations of Ice Nucleating Particle Concentrations, European Aerosol Conference 2024, Tampere, Finland, 25-30 Aug 2024."

14. **Section 2.1: I am a little confused by the description of the setup. Are the PCR plates partially immersed in the ethanol in the aluminium block? Or is the aluminium block an enclosure for the ethanol but has a shaped top that allows the plates to be inserted? A photograph of the setup in the SI would be incredibly useful.**

We are sorry for the confusion, the set up corresponds to what the reviewer asked in the second question. The aluminum block is an enclosure for the ethanol but has a shaped top that allows the plates to be inserted. We have included a photograph of the setup in the SI as suggested (Figure S1).

15. **Section 2.2: The authors may want to provide an explanation as to why the grayscale value, after the initial nucleation event, drops and then rises again as this may confuse those unfamiliar with the freezing characteristics of droplets.**

We thank the reviewer for the suggestion. We have added the following information to the text (L159-163):

"After nucleation occurs, the latent heat released during the phase change slows the freezing process, resulting in a continuous change in the optical signal until the droplet is fully frozen. This behavior can produce multiple features in the grayscale signal, corresponding to nucleation and subsequent solidification, as observed in other immersion freezing setups (David et al., 2019)."

16. **Section 2.3: The LAUDA chiller is ultimately used as the temperature reference for all experiments, but there is no description of mention of how the temperature of the LAUDA chiller itself is calibrated or checked. If the Pt100 is checked against the LAUDA chiller annually then this suggests the chiller has been in use for years, and therefore may have experienced a drift in its temperature measurements.**

Sorry for the confusion, the LAUDA chiller (which has a Pt1000 probe) is used only as reference for the temperature characterization of the external Pt100 probes. For the regular experiments of GRAINS to obtain the INP concentration from samples we use the temperature inside the block as reference, and then apply the correction found in Section 2.3 to account for the difference in temperature of the wells of the PCR plates. We have modified the text between L201-217 to make this clearer:

"For temperature characterization, a total of four Pt100 probes were used: one probe continuously monitored the temperature of the aluminum block, while three additional probes (RTD PT100 RS PRO, Cat. No. 262-3278) were positioned in the PCR plate wells to measure the temperature inside the wells. To check the precision of the temperature measurements among sensors we performed a comparison between the four Pt100 probes and the temperature reported by the LAUDA thermostat, measured with a built-in Pt1000 probe. This was carried out by immersing the Pt100 probes in the ethanol bath of the thermostat at 0 ºC for 15 minutes. Then, the stability of the temperature reading at decreasing temperature was studied, to check the accuracy of the probe at the usual temperature range of an experiment. For that, we again introduced the Pt100 sensors in the ethanol bath and measured at seven different temperatures from 0 ºC to -30 ºC in steps of 5 ºC. At each temperature, we recorded the temperatures measured by the Pt100 probes and the one reported by the LAUDA thermostat for 5 minutes. The results showed an excellent agreement, following a 1:1 relationship for the entire temperature range. This intercomparison is performed annually to ensure a correct reading of the temperature in the experiments.

Then, to characterize the temperature measurements in GRAINS, we established a relationship between the temperature of the aluminum block measured by the main Pt100 sensor and the temperature inside the wells of the PCR plates. For that, we used the three extra Pt100 probes that have dimensions of 2x10 mm and are small enough to be completely introduced in the wells."

Regarding the calibration of the LAUDA chiller, it is not possible to access its component to perform a calibration of the sensor. Instead of that, we intercompare the Pt1000 chiller sensor and the Pt100 sensors and we check that the differences are within the error of the probes. The chiller was acquired in late 2023, so it has only been used for a bit more than two years, and hasn't shown any deviations when compared to the other Pt100 probes, so we trust that there is no drift in its temperature measurements.

17. **Line 203: The gap between the aluminium block and the plates is described as being filled with ethanol here, but this is not mentioned during the description of the standard operation of GRAINS, so is not an accurate representation of the temperature of the wells in relation to the aluminium block during a typical droplet freezing assay – I now see that this is addressed at the end of section 2.3, but this part of the procedure (adding ethanol between the plate and the block) should be included in Section 2.1 too since it is part of the standard procedure.**

We have added this information in Section 2.1 (L136-137) as suggested by the reviewer.

18. **Figures S1 and S2: Given the wide spread in temperatures between the wells and blocks depending on position (assuming these are reproducible, I do not see any uncertainty values, presumably for clarity), could the temperature uncertainty be reduced by having bespoke temperature corrections for each well rather than a "blanket" correction?**

The reviewer made a very interesting point. We performed the temperature correction experiments in the wells shown in Figure 1 because we considered that they were a first accurate representation of different positions inside the PCR plate and represented different distances from the point where the ethanol enters the aluminum block. Our first idea was to find a function for temperature correction (based on the experiments performed) that could be applied to all wells in the PCR plate. However, we did not see any trend in the temperature difference depending on the region of the PCR where the experiments were performed, and the standard deviations of these temperature differences are within the error of the Pt100 probes used for the experiments. In fact, the mean temperature differences found for both PCRs were practically identical. For these reasons, we finally decided to apply just one correction for all the wells in both PCR plates.

19. **Line 260: There should be a space between the "g" and "L-1" in the units of g L-1. Likewise between the m2 and g-1 in line 261, i.e. m2 g-1. I also see this throughout the manuscript, all units should have spaces between the different components of the units.**

Thank you for the note, we have corrected all units throughout the manuscript.

20. **Line 271: Please note that some surface area will not be accounted for since particles smaller than 0.5 um are not measured.**

We have added the following sentence to the text (L287-288): "However, due to the measurement range of the APS, the surface area of particles below 0.5 µm in diameter will not be accounted for in S_{total}."

21. **Line 278: Are the original sample and dilutions all performed in one GRAINS experiment by distributing them across the two plates? Or are they run one after another? How many repeats were performed?**

For the NX Illite experiments we used one PCR plate for each suspension (original sample and two dilutions), so they ran one after another in each set of experiments. We performed four sets of experiments for the dry dispersion method and three sets of experiments for the wet suspension method. Results of the individual spectra for each set of experiments are shown in Figure S7.

22. **Line 318: What particular sample of K-feldspar was used, e.g. FS02?**

The K-Feldspar used was the BCS-CRM No. 376/1 POTASH FELDSPAR SGT FELDSPAR 1, we have added the abbreviation "BCS-CRM 376/1" to the description of the samples used in the AIDAd experiments in L336.

23. Line 347: The collection of field blank filters is excellent practice. Was this also done for the other filter measurements described in the previous sections?

Unfortunately, we did not collect blank filters for the other sets of experiments. We performed another campaign at AIDAd (not included in this manuscript) and this time we analyzed blank filters. Results showed that the freezing activity of the blank filters were very similar to the freezing activity of ultrapure filtered water, so we are confident that this would be the same for the intercomparison campaign, as the protocol is the same. On the other hand, the filters used for the NX Illite experiments are equivalent to those used for the comparison of methodologies, so we would also expect the same results for the blank filters in these experiments.

24. Line 365: Clarify that the punched filter parts were added to each well, it is implied but not actually stated.

Thank you for the suggestion, we have included this information (L387).

25. Lines 368-370: Can the authors clarify why this method was employed? Is it a standard method used in the literature, in which case can citations be added (as for the other filter techniques)? For a more direct comparison to polycarbonate filters, could the quartz filters also be washed as a full filter (i.e. without punching)? Or were the 1 cm punches taken from the same filter as the other 1 mm punches?

We decided to employ this method (Quartz punch washed) because we considered that the process in the laboratory gets simplified compared to the Quartz 96-punch, where the operator has to perform 96 individual punches and introduce them into each well of the PCR plate, which can be very time consuming. Furthermore, since the sample is exposed for a longer time, contamination due to handling is more likely to occur. Additionally, we have tested the performance of dilutions with the Quartz punch washed method and it worked as well as with the Polycarbonate method, so it allows to extend the range of INP concentrations obtained with droplet freezing arrays. There are not numerous studies that use this method; we only found data from LINDA (LED-based Ice Nucleation Detection Apparatus; Stopelli et al., 2014) in Bras et al. (2024) and Lacher et al. (2024). Therefore, we considered adding it to the comparative of methodologies and test if it could potentially become an alternative to the Quartz 96-punch method. As suggested, we have added the citations into the text (L393-394).

We believe that washing the whole filter in water would not be very practical, since it would require a large amount of water given the diameter of the filter (150 mm). Also, as it will be further discussed in comment 42, the quartz filter is mainly used for chemical analysis, so in our case it would not be possible to wash it completely. In fact, we believe that a key advantage of using quartz filters is that both the chemical composition and the INP concentration can be obtained from the same filter sample. Here, we cut one quarter of the filter while the remaining 3/4 of the filter are used for chemical composition determination. In the 1/4 piece of filter, we performed the 1 cm punch and the 96 1 mm punches for INP analysis. So, to answer the reviewer question, yes, the 96 punches were done in the same filter as the 1 cm punch.

26. Line 373: "where *V*punch is the volume of air that passed through one punch of the filter" – is this calculated as a fraction of the total amount of air that passed through the full filter? In which case, does this also take into account the "unused" portion of a filter where the O-ring is seated?

Yes, the reviewer is right and V_{punch} is calculated as the fraction of the total air that passed through the entire filter. However, we were indeed considering the part of the filter where the O-ring is seated. In the revised version of the manuscript, we have only considered the part of the filter that was actually sampled for the calculations of V_{punch}, which we believe is more accurate. This change implies a 20% difference in the INP concentrations calculated with both Quartz punch methods, so we have updated Figure 6 in the revised version of the manuscript, which we show here as Figure R4.

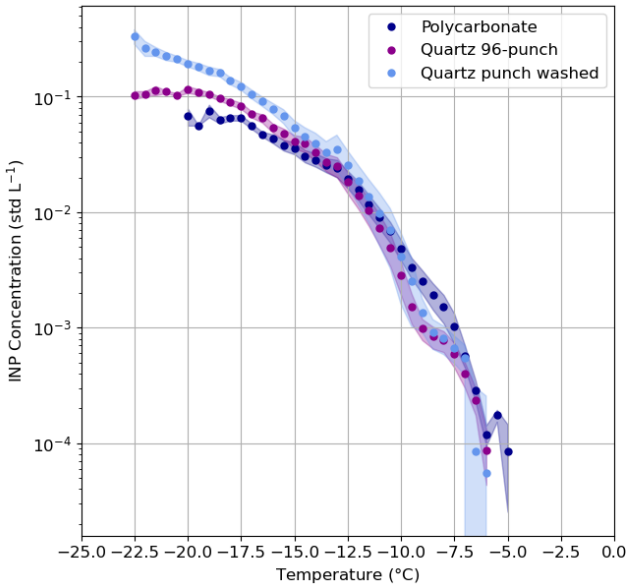

Figure R4. Comparison of the different methods for sampling and analyzing INP. Data shown correspond to the average of the individual spectra. Shaded area represents the standard error of each method.

27. When analysing multiple 1 mm punches with GRAINS, are the freezing patterns consistent with homogeneous dispersion of aerosols across the filter surface? Or can discrepancies be seen, e.g. stronger signal in punches taken from the centre of the filter compared to nearer the edge?

This is a very interesting point. We assume a homogenously sampled filter for INP and chemical analysis, so the different portions of the filter undergo different analysis that are later on combined under the assumption of homogenous sample distribution. In our protocol, what we do is to visually inspect the filter in case obvious inhomogeneities exist prior to cutting it.

In particular, since for the INP analysis the punches were taken randomly, it is not possible to determine a relationship between freezing patterns and region of the quartz filter where the punch was taken from. It would be interesting to test whether this has an effect or not during a dedicated experiment. Concerning the Quartz punch washed method, we tested its variability by evaluating the INP concentration obtained from different 1 cm punches in one quartz filter. Figure R5 shows that we obtain similar INP concentrations that are within uncertainties, so the assumption of a homogeneous dispersion of aerosol particles across the filter can be considered valid at that scale probed by the 1 cm punches.

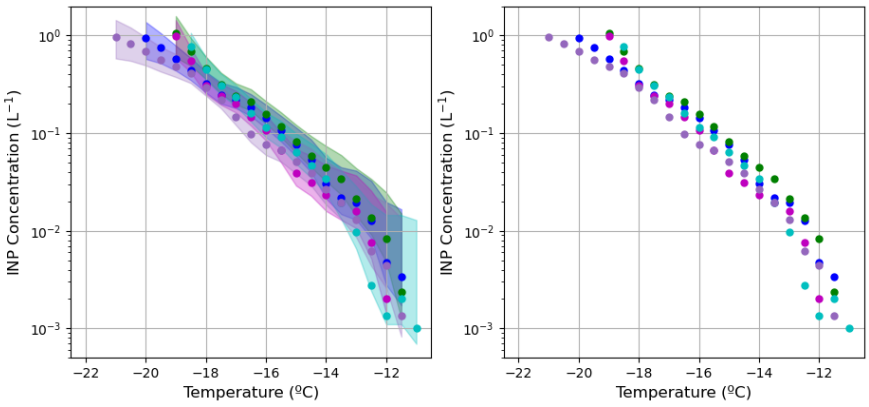

Figure R5. INP spectra obtained following the Quartz punch washed method for different regions of one quartz filter with uncertainties by Agresti and Coull (1998) (left panel) and without them (right panel).

28. Line 390: It is mentioned in a comment above, but without a measurement of the population of smaller particles, e.g. with an SMPS, how confident are the authors of their total surface area measurements.

We are confident of our surface area measurements since we measured the complete size distribution of our NX Illite sample (SMPS + APS) at AIDAd but after a 2.5 μm impactor, evidencing low contribution of particles below the APS lower size limit. To estimate the contribution of the surface area from smaller particles, we have compared both surface size distributions, the one measured at AIDAd and the measurements from section 4.1 (which are shown in Figure S6 of the SI), normalized by concentration of particles. We then calculated the ratio between the surface area of the fine mode measured by the SMPS (at AIDAd) compared to the one used for the calculation of n_s in Figure 4. The ratio obtained was 0.045, so the smaller particles would not contribute much to the total surface area. Considering the contribution of the fine mode would imply a 4.5% decrease in the calculated n_s, which is within the uncertainties derived from Agresti and Coull (1998).

29. Figure 3: The authors might also consider including the WSIDOM data from Reicher et al. 2018: https://amt.copernicus.org/articles/11/233/2018/. Further, it may be useful to show the droplet volume for each technique in the legend since GRAINS uses a very large volume compared to most other immersion freezing techniques shown here.

We appreciate the note, but unfortunately the WISDOM data are not publicly available, and requesting the data to the authors would delay the review process of the manuscript. We believe that Figure 3 has a considerable number of datasets for comparison and the figure is already a bit loaded, so adding new datasets would further complicate the visualization. On the other hand, we have included droplet volume (or radius) for each device when available.

30. Figure 3 caption: Mention that A13 is for K-feldspar. Also, it is not entirely clear in the text why A13 is shown; is the scaling factor applied here for A13 related to the amount of K-feldspar found in NX illite? Is there a reference for this?

Thank you for the note, we have added this information to Figure 3 caption. Yes, this scaling factor was proposed by Hiranuma et al. (2015). As there was a 14% of feldspar in their NX Illite sample, they scaled the A13 parametrization by multiplying for a factor of 0.14. However, this parametrization overestimated the n_s by NX Illite, so they lowered the factors used to be in the range of the found n_s values. We followed a similar approach here, and applied lower scaling factors to be in the range of the obtained values.

31. Figure 3 caption: The authors state that uncertainties are as per Agresti and Coull (1998), but there is no indication of what these uncertainties are or how they are calculated. Further, there is no citation for Agresti and Coull in the reference list.

Sorry for the mistake, the reference is now included in the reference list (L840-841). Agresti and Coull (1998) proposes a method for calculating the confidence intervals for the approximation of a binomial distribution. Since the frozen fraction follows a binomial distribution, one can calculate the confidence intervals following Eq. 2 in Agresti and Coull (1998):

$$\frac{p + \frac{z_{a/2}^2}{2n} \pm z_{a/2} \cdot \sqrt{\left(\frac{p(1-p) + \frac{z_{a/2}^2}{4n}}{n}\right)}}{1 + \frac{z_{a/2}^2}{n}}$$

where p is the sample proportion (which corresponds to the frozen fraction), n is the sample size (which corresponds to the number of droplets), a is the confidence level (0.05 for 95%) and $z_{a/2}$ is the critical value of the standard normal distribution (which is 1.96 for the 95% confidence interval).

We have followed this approach for the frozen fraction. Then, uncertainties in derived variables were obtained by evaluating the analytical expressions at the lower and upper confidence limits of the frozen fraction. We have explained this in L434-436:

"Uncertainties for the frozen fractions were calculated from the 95% confidence interval for binomial sampling, following Agresti and Coull (1998), whereas uncertainties in derived variables were obtained by evaluating the analytical expressions at the lower and upper confidence limits of the frozen fraction."

Agresti, A. & Coull, B. A. (1998). Approximate is Better than "Exact" for Interval Estimation of Binomial Proportions, Am. Stat., 52:2, 119-126. https://doi.org/1010.1080/00031305.1998.10480550

32. Figure 4: The authors should comment on some apparent systematic biases since the comparisons to FrESH are above the 1:1 line while those with INSEKT are below the 1:1 line.

The reviewer is right, most of the GRAINS-FrESH intercomparison is above the 1:1 line. From Figure S8, one can see that the INP spectra of both devices is practically identical, but the differences in INP concentration at each temperature come from a notable shift in temperature. Since FrESH does not apply yet any correction to the temperature in the wells, we believe that this might be the cause of the differences in INP concentration values, since both devices measure very similar curves. We have added some comments about this in the GRAINS-FrESH intercomparison (L496-506):

"The GRAINS-FrESH intercomparison shows that the first device tends to measure higher INP concentrations at the same temperature. In fact, there is one sample in the GRAINS-FrESH intercomparison that shows substantial differences between both instruments, KUO20231001, being the INP concentrations obtained with GRAINS around one order of magnitude higher than those obtained with FrESH. Figure S8, that contains the individual GRAINS and FrESH INP spectra obtained from each sample, shows that the behavior in the INP concentration spectra obtained with both devices is mostly the same, but with a shift in temperature, which may be due to the fact that FrESH does not yet account for a temperature correction of the wells of the PCR plates. In general, there is an increased scattering of the data for lower INP concentrations, while the opposite occurs for higher INP concentrations, where most of the data is closer to the 1:1 line. For these samples, Figure S8 shows that the behavior of the freezing spectra obtained with the two droplet freezing arrays is quite similar and this temperature shift is less pronounced."

Concerning INSEKT, we stated the possible reason for larger differences observed in Figure S9 due to one of the dilutions used in the experiments. However, since these higher values obtained with INSEKT are only noticeable when analyzing the ambient samples (for the intercomparison at AIDAd the INP concentrations are very similar), we believe that is not a systematic bias.

33. Section 4.2: This might work better divided into sub-sections for each of the sets of intercomparisons.

We appreciate the suggestion. After revisiting this section, we believe that dividing it into two subsections (one for ambient samples and one for aerosol standards) wouldn't significantly improve the flow. Clear distinctions between the two sets of experiments are already made, and we believe the section reads smoothly as written.

34. Figure 5: The large discrepancy between GRAINS/INSEKT and PINE is concerning. Are there any previous studies of K-feldspar with PINE during other intercomparisons to check whether this is a common problem? That GRAINS and INSEKT compare well is good.

We agree with the reviewer, this is something that has to be investigated in more detail. Our hypothesis is that the droplet freezing arrays might be underestimating the INP concentration of such a highly active substance because of the assumption of a single INP per droplet (Vali, 1971). Another hypothesis could be that the ice nucleation activity of K-feldspar is degraded due to being immersed in water.

For further details on this PINE vs INSEKT-GRAINS comparison, we refer the reviewer to the response to their major comment, where we explained in more detail the potential reasons why there is an underestimation of the ice nucleation ability of K-feldspar with GRAINS and INSEKT.

35. Section 4.2: Given that there is a lot of literature data available for K-feldspar, I am surprised to see no literature comparisons here. Can the authors provide some comparisons to the literature data? This would actually fit better with Section 4.1. The same could also be done with ATD.

Following the reviewer's suggestions, especially those provided in their major comment, we have included a comparison of the ice nucleation properties of the four dust samples and existing parametrizations in the literature, which can be found between L565-585 in the revised manuscript. Also, we have updated Figure S10 from the SI (Figure R6 in this document) and it now includes the parametrizations from the literature. However, we have kept this information in Section 4.2, to be consistent with the subsections in the methodology.

L565-585: "Figure S10 also shows parametrizations reported in the literature for ATD (Niemand et al. 2012 – N12), K-Feldspar (Atkinson et al. 2013 – A13) and dust (Niemand et al. 2012 – N12; Ulrich et al. 2017 – U17). In general, the slopes of the n_s spectra for ATD, SDSA01 and Saharan dust follow the same trend as the dust parametrizations, but the experimental data shows slightly lower values of n_s, especially for both droplet freezing arrays. For K-Feldspar the two spectra overlap for the majority of the temperature range, but PINE n_s values are larger than the ones obtained with the two droplet freezing arrays, by more than two orders of magnitude. In fact, PINE data shows n_s values similar to those corresponding to the A13 parametrization, whereas INSEKT and GRAINS show the same slope of the spectra as the parametrization by Atkinson et al. (2013), but with a difference of two order of magnitude in the values. This could be due to the fact that the working principle of INSEKT and GRAINS assumes that there is a single INP per droplet in the PCR wells (Vali, 2019), and since K-Feldspar is a highly ice-active substance the n_s obtained with both droplet freezing arrays could be underestimated. Another possibility could be that the ice nucleation activity of K-Feldspar is degraded due to being immersed in water. This is something observed by Harrison et al. (2016), although the degradation occurred after the sample was immersed in water for months. Also, Kumar et al. (2018) stated that when K-Feldspar is immersed in water there are interactions between the water and the sample's surface, which could explain the difference between the ice nucleation ability obtained with droplet freezing arrays compared to PINE and the A13 parametrization. Apart from the case of K-Feldspar, the results shown in Figure 5 and Figure S10 confirm the similarity of the two droplet freezing arrays and the PINE instrument, confirming that GRAINS can reproduce the freezing behavior of mineral dust samples when compared to INSEKT and PINE."

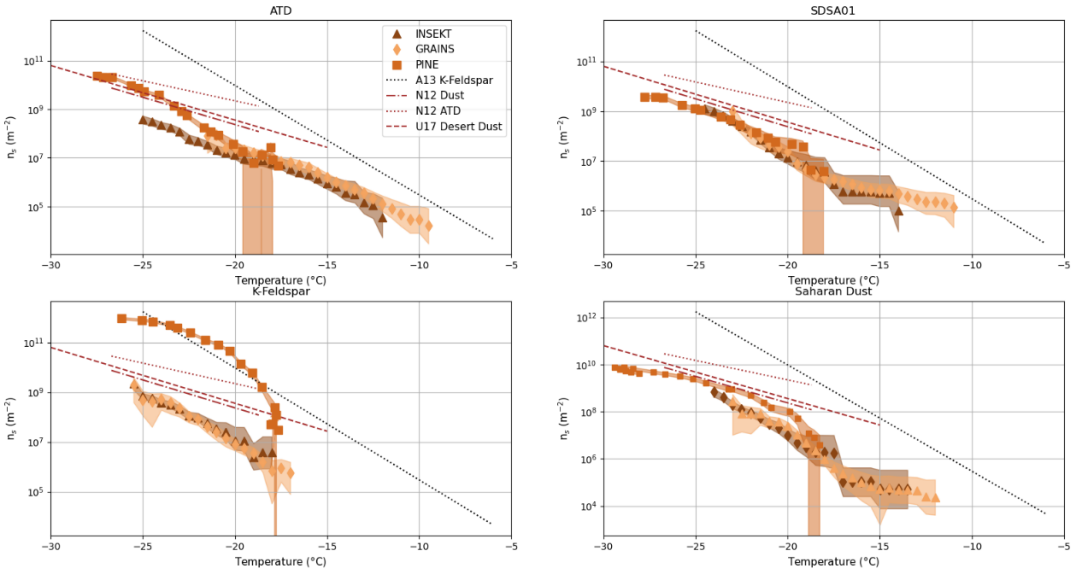

Figure R6. Ice nucleation active sites (INAS) density (n_s) per unit of aerosol surface obtained with INSEKT, GRAINS and PINE during the experiments conducted at AIDAd. Surface area was obtained as the lognormal fit of the size distributions obtained with the combination of the Scanning Mobility Particle Sizer (SMPS) and Aerodynamic Particle Sizer (APS). Shaded area represents uncertainty following error propagation, where the error of the frozen fractions for INSEKT and GRAINS was calculated based on Agresti and Coull (1998), whereas for PINE it was calculated as a combination of a 10% of the INP concentration and the square root of the number of ice crystals. Parametrizations from Atkinson et al. (2013) (A13), Niemand et al. (2012) (N12) and Ulrich et al. (2017) (U17) are also shown.

36. Section 4.3: Can the authors explain why different cut-offs were used for the filter samplers during this comparison? That they are not 1:1 suggests there could be differences in results, although this may only affect the "tail ends" of the data rather than the "main body" of the INP curves.

We understand the concern and agree that for a better comparison the sampling conditions (i.e., cut-offs) should be the same. However, quartz filters are regularly sampled at our measurement station for chemical analysis purposes using a high-volume sampler with a PM10 head. On the other hand, the main aerosol inlet at the station where the in-situ instruments are connected is a whole air inlet with no aerosol cut-off. We connected a custom-made low-volume sampler, the GRAnada Sampling System (GRASS), to this whole air inlet for sampling the polycarbonate filters. This way, these measurements and the rest of in-situ measurements (aerosol optical properties and size distribution) are performed under the same conditions.

That said, we confirmed with measurements of the aerosol size distribution from the APS, which is connected to the whole air inlet as well, that the concentration of particles larger than 10 µm is very low. Therefore, the inlet cut-off is expected to have a minor impact on the comparison. Furthermore, as the reviewer stated, this would not affect the INP curves much, although it is still a potential source of discrepancies in the results that cannot be ruled out at all.

Finally, we would like to emphasize that the purpose of this comparison is to explore the potential of using routine PM10 sampling at air-quality monitoring stations to derive INP data from the same filter sample. Therefore, despite differences in size cut-off, the results presented here demonstrate promising prospects for this approach.

37. Figure 6 and lines 553-555: The authors use standard error of the mean here, whereas throughout the rest of the paper they calculate the error based on Agresti and Coull (which

is never explained). There should be consistency in treatment of the uncertainties throughout the manuscript and dataset.

Yes, we have used the standard error here because we calculated the average curve of all INP concentration spectra, so we consider that the error should reflect the variability of the curves involved in the average rather than the measurement uncertainty. As for Agresti and Coull, we have explained the source of this method in comment 31.

38. Lines 571-572: Does Figure 6 shows the average of all 27 samples? Or an individual sample? I am very surprised to not see the data for the 27 samples in the SI, I feel that should be included.

Figure 6 shows the average of all 27 samples. The reviewer is completely right about the data for the individual samples. Figure R7 shows the individual spectra for the comparison of the different methods for sampling and analyzing INP. As can be seen, there is some variability in the comparison between samples, which is reflected in the standard error of the mean reported in Figure 6. We have included this figure in the SI as Figure S11. Please note that in this case we have not included uncertainties for clarity reasons.

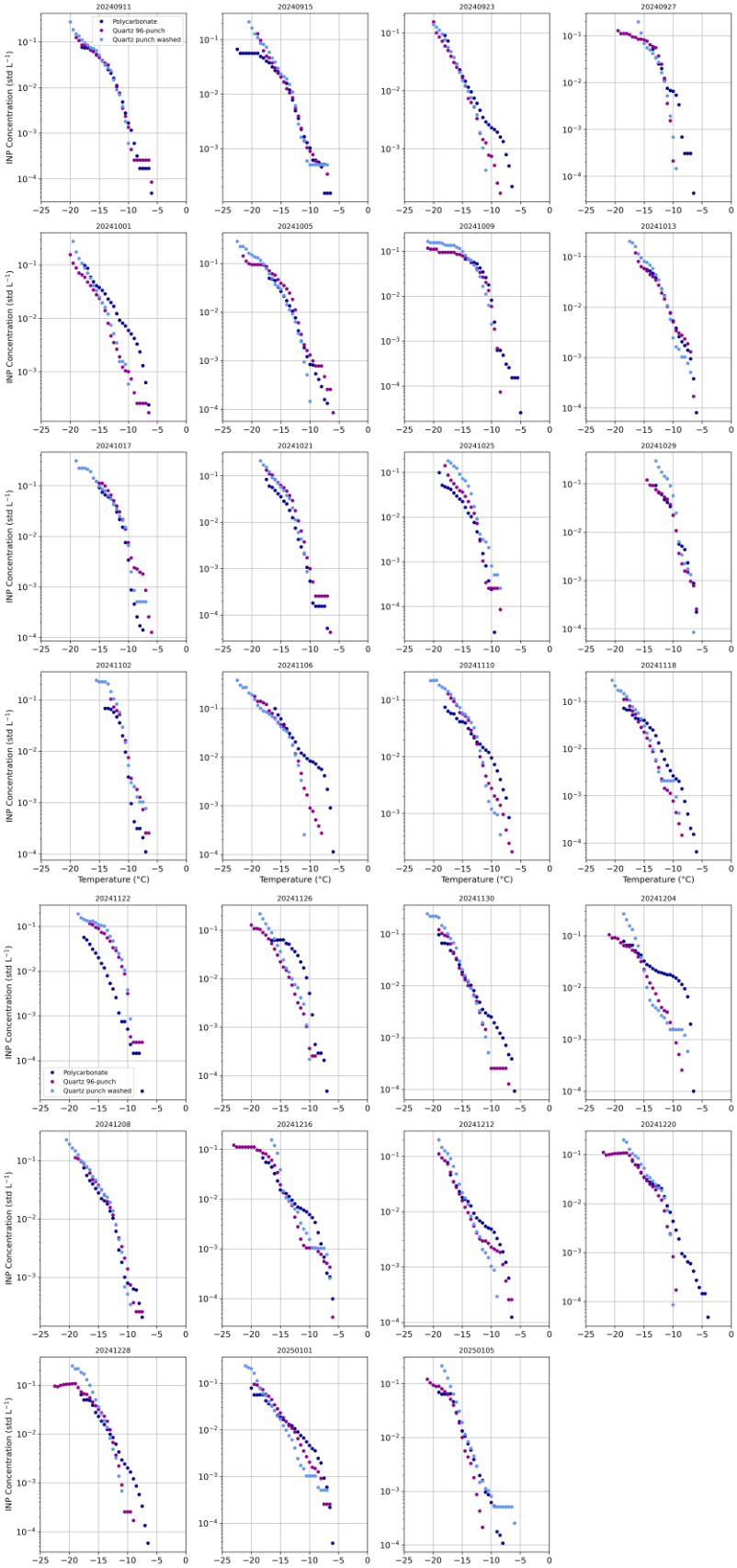

Figure R7. Individual spectra for the comparison of the different methods for sampling and analyzing INP. Uncertainties are not shown for clarity.

39. Section 4.3: An issue with this set of experiments is that the aerosol composition is unknown since they are ambient samples. Therefore, it is not possible to say whether differences in INP concentrations at temperatures colder than around -13 oC could be due to the differences in collection efficiency of different types of INPs across the sampling techniques. This comparison would have been better incorporated into controlled lab studies such as those in Section 4.2.

We agree with the reviewer that using aerosol of known composition helps to rule out other factors affecting the comparison. However, we also believe that the number of samples and the varying aerosol conditions during the 3-month sampling period allow us to obtain robust results. As discussed in comment 2, the sampling period included days dominated by anthropogenic pollution, background conditions as well as days affected by transported Saharan dust, and the differences observed do not seem to be related to the different atmospheric scenarios. In L631-656 we included a discussion on the differences between INP concentration with each method depending on the aerosol conditions at the station.

L631-656: "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."

40. Line 641: "detail" rather than "detailed".

Thanks, we have corrected the typo.

41. Line 642: This begs the question of why this wasn't done as part of the AIDAd intercomparison described earlier.

Unfortunately, the comparison of different filter substrates started prior to the AIDAd campaign, and we made use of the instrumentation available at the UGR station. We would like to highlight that the AIDAd facility is highly demanded, so for the campaign we focused on the GRAINS-INSEKT intercomparison as part of the validation of GRAINS. We believe that future experiments evaluating the differences in filter substrates and extraction methods under controlled laboratory conditions are necessary and could be a future work of the authors to be performed at the AIDAd facility.

Nevertheless, we consider that an ambient comparison is also very helpful, since it reflects the actual sampling conditions of quartz filters across different stations worldwide.

42. **Line 644-645: Rather than punching a 1 cm hole, could the entire filter not be washed as in the polycarbonate method, thus reducing preparation further? Or is the rest of the quartz filter used for further analyses?**

As stated in comment 25, the quartz filter is used for chemical analysis, so it would not be possible to use it completely for the Quartz punch washed method. Here, we cut one quarter of the filter and from that section we use the 1 cm punch and the 96 1 mm punches for INP analysis.

43. **Line 675: The authors should reiterate here that there is a discrepancy with K-feldspar between the PINE and the cold stage instruments that warrants further study.**

We have included the following information in the conclusions (L771-774):

"In this sense, the n_s of K-Feldspar differed in two orders of magnitude between PINE and the droplet freezing arrays, probably because the offline methods underestimate the ice nucleation ability of this highly active sample. Still, further study is necessary to understand these differences."

44. **What is the nominal pore size of the quartz filters? Could their capture efficiency of smaller particles be greater than for the polycarbonate filters, which could potentially help to explain some of the discrepancies are colder temperatures?**

Since quartz filters consist of a matrix of quartz fibers that overlap, there is not a defined pore size such as in the polycarbonate filters. Particularly we use Pallflex filters (Pall), and the manufacturer guarantees a 99.9% aerosol particle retention.

45. **Line 706: Do the authors mean "immersing" or "washing" rather than "dissolving"?**

Yes, thank you for the note.

46. **Lines 707-710: This is an excellent point and should be reiterated in the discussion of Section 4.3.**

We thank the reviewer for their comment, we had included this discussion at the end of Section 4.3 in the first version of the manuscript. We have added more information to the section, in particular between L734-739:

"Therefore, the methods are clearly related, with a slightly higher positive correlation between the Quartz 96-punch method and the Polycarbonate method. Again, these differences between the substrates need to be investigated in more detail, by sampling well-known particles that are characterized in composition and size. This would potentially allow to extend the existing INP database by using standard PM10 quartz filters that are commonly and regularly sampled at many atmospheric observatories, allowing a more comprehensive characterization of atmospheric ice nucleation in different environments."

And also between L669-671:

"Therefore, the three methods show good agreement between each other, indicating that quartz filters, typically used for chemical analysis, could potentially be used to extend global coverage of INP measurements."

References:

Atkinson, J. D., Murray, B. J., Woodhouse, M. T., Whale, T. F., Baustian, K. J., Carslaw, K. S., Dobbie, S., O'Sullivan, D., & Malkin, T. L. (2013). The importance of feldspar for ice nucleation by mineral dust in mixed-phase clouds. *Nature*, 498(7454), 355–358. https://doi.org/10.1038/nature12278

Bras, Y., Freney, E., Canzi, A., Amato, P., Bouvier, L., Pichon, J., Picard, D., Minguillón, M. C., Pérez, N., & Sellegri, K. (2024). Seasonal Variations, Origin, and Parameterization of Ice-Nucleating Particles at a Mountain Station in Central France. *Earth and Space Science*, *11*(6). https://doi.org/10.1029/2022EA002467

Conen, F., Henne, S., Morris, C. E., & Alewell, C. (2012). Atmospheric ice nucleators active ≥ −12 °C can be quantified on PM₁₀ filters. *Atmospheric Measurement Techniques*, *5*(2), 321–327. https://doi.org/10.5194/amt-5-321-2012

Harrison, A. D., Whale, T. F., Carpenter, M. A., Holden, M. A., Neve, L., O'Sullivan, D., Vergara Temprado, J., and Murray, B. J. (2016) Not all feldspars are equal: a survey of ice nucleating properties across the feldspar group of minerals, *Atmos. Chem. Phys*., 16, 10927–10940, https://doi.org/10.5194/acp-16-10927-2016

Kumar, A., Marcolli, C., Luo, B., and Peter, T. (2018), Ice nucleation activity of silicates and aluminosilicates in pure water and aqueous solutions – Part 1: The K-feldspar microcline, *Atmos. Chem. Phys*., 18, 7057–7079, https://doi.org/10.5194/acp-18-7057-2018

Lacher, L., Adams, M. P., Barry, K., Bertozzi, B., Bingemer, H., Boffo, C., Bras, Y., Büttner, N., Castarede, D., Cziczo, D. J., DeMott, P. J., Fösig, R., Goodell, M., Höhler, K., Hill, T. C. J., Jentzsch, C., Ladino, L. A., Levin, E. J. T., Mertes, S., Möhler, O., Moore, K. A., Murray, B. J., Nadolny, J., Pfeuffer, T., Picard, D., Ramírez-Romero, C., Ribeiro, M., Richter, S., Schrod, J., Sellegri, K., Stratmann, F., Swanson, B. E., Thomson, E. S., Wex, H., Wolf, M. J., and Freney, E. (2024), The Puy de Dôme ICe Nucleation Intercomparison Campaign (PICNIC): comparison between online and offline methods in ambient air, *Atmos. Chem. Phys*., 24, 2651–2678, https://doi.org/10.5194/acp-24-2651-2024

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

Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., & Leisner, T. (2012). A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles. *J. Atmos. Sci.*, 69(10), 3077-3092. https://doi.org/10.1175/JAS-D-11-0249.1

Stopelli, E., Conen, F., Zimmermann, L., Alewell, C., & Morris, C. E. (2014). Freezing nucleation apparatus puts new slant on study of biological ice nucleators in precipitation. *Atmos. Meas. Tech.*, *7*(1), 129–134. https://doi.org/10.5194/amt-7-129-2014

Tatzelt, C., Henning, S., Welti, A., Baccarini, A., Hartmann, M., Gysel-Beer, M., van Pinxteren, M., Modini, R. L., Schmale, J., & Stratmann, F. (2022). Circum-Antarctic abundance and properties of CCN and INPs. *Atmos. Chem. Phys.*, 22(14), 9721–9745. https://doi.org/10.5194/acp-22-9721-2022

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. *Atmospheric Environment*, *89*, 593–602. https://doi.org/10.1016/j.atmosenv.2014.03.001

Ullrich, R., Hoose, C., Möhler, O., Niemand, M., Wagner, R., Höhler, K., Hiranuma, N., Saathoff, H., & Leisner, T. (2017). A New Ice Nucleation Active Site Parameterization for Desert Dust and Soot. *J. Atmos. Sci.,* 74(3), 699-717. https://doi.org/10.1175/JAS-D-16-0074.1

Welti, A., Müller, K., Fleming, Z. L., & Stratmann, F. (2018). Concentration and variability of ice nuclei in the subtropical maritime boundary layer. *Atmos. Chem. Phys*, 18(8), 5307–5320. https://doi.org/10.5194/acp-18-5307-2018

Wex, H., Augustin-Bauditz, S., Boose, Y., Budke, C., Curtius, J., Diehl, K., Dreyer, A., Frank, F., Hartmann, S., Hiranuma, N., Jantsch, E., Kanji, Z. A., Kiselev, A., Koop, T., Möhler, O., Niedermeier, D., Nillius, B., Rösch, M., Rose, D., Schmidt, C., Steinke, I., and Stratmann, F. (2015). Intercomparing different devices for the investigation of ice nucleating particles using Snomax® as test substance, *Atmos. Chem. Phys*., 15, 1463–1485, https://doi.org/10.5194/acp-15-1463-2015

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. (2019). Annual variability of ice-nucleating particle concentrations at different Arctic locations. *Atmos. Chem. Phys,* 19(7), 5293–5311. https://doi.org/10.5194/acp-19-5293-2019

Vali, G. (1971). Quantitative Evaluation of Experimental Results an the Heterogeneous Freezing Nucleation of Supercooled Liquids. *J. Atmos. Sci.*, *28*(3), 402–409. https://doi.org/10.1175/1520-0469(1971)028<0402:QEOERA>2.0.CO;2