

We would like to thank anonymous Referee #2 for his/her constructive suggestions, which helped us to improve the manuscript. Specific answers are given in blue text and manuscript modifications related to the Reviewer's comments are given in green text. Line numbers below correspond to the clean revised manuscript.

Report #1 Second Round, Anonymous Referee #2:

2nd round of review of „Ice Nucleating Particles Variability Across a Megacity“ by Sebastián Mendoza-Téllez et al., submitted to EGU sphere.

The manuscript did improve much, in general. However, there are still two major issues that do not allow for publication in its current form. Indeed, the current way of depicting results got worse for these two topics, compared to the first version. I describe and discuss these issues below and give suggestions for improvements. There are a few additional specific comments, together with a list of simple corrections.

Should the authors be able to improve these points of concern, the work can be published. But in any case, I leave it to the editor to decide if this should be rather a "measurement report", as the authors say they are open to such a move, and as I see the work somewhere in between "article" and "measurement report".

A/ Thank you for the new suggestions and recommendations. Please find our specific answers below.

1) As you can see yourself in your Equation 2, there is a range of INP concentrations [INP] you can get:

You say that you observe roughly 30-40 droplets, so let's assume 35 on average. With that, the highest and lowest value that the "In-part" in Equation 2 can take on values between 0.029 and 3.56. All other parameters, I denoted as "X" below.

$$[INP(T)] = -\ln(Nu(T)/No) \cdot X$$

X depends, among other parameters, on the volume of air sampled. The maximum and minimum values that [INP] can take on therefore varies proportional with X. If you sample a 10 times higher volume, compared to your original measurement, the lowest [INP] you obtain is 10 times lower than for your original measurement. With this, T_0 and T_50 will be much higher. Therefore, T_0 and T_50 are not useable, UNLESS sampling and INP measurements were done in an identical way.

This is also true for other off-line techniques for INP measurements. There also the droplet size plays a role, together with the sampling volume and other parameters. Even if the identical sample was examined in two off-line techniques which each examines the same number of droplets of 1 and 50 microliter, the latter will have a detectable concentration range that is 50 times lower than the former. And with that, both T_0 and T_50 will be different (i.e., higher for the latter).

For an INP spectrum with a typically observed slope of -0.5 (taken from Fletcher, 1962), a factor of 10 or 50 in [INP] means a delta in T_0 and similarly in T_50 of 4 and 7K, respectively! Check it for yourself. As your curves seem flatter than that from Fletcher (1972), likely the difference between your results and those from others who did not measure in the exact same way (including same sampling volumes) may even be larger.

This makes any interpretation of T_0 and T_50 meaningless.

You may argue that you could use it to compare between your own results. But there, differences in the measurement range for the 10 INP spectra (original Fig. 3, and also the difference in range

between the two curves of the actual Fig. 3) already show that even for these the sampling and measurement conditions were not exactly the same.

Also, for different MOUDI stages, data from different stages should be expected to be different: larger particles are more ice active (due to the larger surface area, as you said), but likely fewer large particles are there. So any results can follow, with higher or lower T₀ and T₅₀ between different stages, depending on the particle mix. And while you may show these values, you should not average those from different stages.

Long story short: It would be best if you used only INP concentrations throughout. This mainly refers to Fig. 2 and all related parts of the text.

A/ Thank you. Following your suggestions we have removed Figure 2 and its corresponding discussion, focusing on INP concentrations only.

If you insist on keeping Fig. 2 as it is now, it needs to be explicitly stated that

a) T₀ and T₅₀ values can only be compared if sampling and measurement conditions between the compared samples were fully the same,

b) T₀ and T₅₀ values are not expected to be the same between different size ranges, but separate sampling sites can roughly be compared because of very similar sampling and measurement conditions were used at both,

c) the boxplots in Fig. 2 need to go, as they average over values that, as explained above, cannot be expected to show the same,

d) the comparison with literature based on T₀ or T₅₀ all has to go.

A side note: Fig. 2(b) and lines 439-442: The circles are difficult to see in Fig. 2(b). If you want to keep Fig. (2) and this description, please make the circles stand out in some way (plot them larger and / or make them bold).

A/Thank you for highlighting out this important point about T₀ and T₅₀. Following your suggestion, we decided to put out all T₀ and T₅₀ results (figures and all related parts of the main text) and use INP concentrations only throughout. We hope that the new revised version covers all your concerns. Figure 2 was deleted.

2) Fig. 3 in the original version showed all 10 INP spectra, 5 each from the northern and southern site. This was the one location where the reader could judge the similarity of 9 of the 10 curves and clearly see that there was one outlying curve.

The authors now decided to remove this one curve. Instead, in the new Fig. 3(a) only the two curves from that one day with the outlier was shown. This is misleading. At the same time, a second panel was added, Fig. 3(b), which still shows the misleading mean values for which all 5 samples were averaged (see my first review).

In my understanding, this is not a good way of dealing with this issue. My suggestion is, to show the original Fig. 3, with all 10 curves, but without the literature data, as a new Fig. 3(a). Fig. 3(b) then could be this same figure, with all data obtained in this study in one color (e.g. grey) in the background, and all the literature data in the foreground, without any averaging or temperature binning. This will clearly show your one outlying INP spectrum, the good comparison with literature, and it will remove all the misleading issues. Related text will need to be revised accordingly.

A/ It seems like we misunderstood your initial idea, we are sorry for this and we thank you for clarifying this key point. We have adjusted Fig. 3 (now presented as Fig. 2), following your advice.

We hope the new revised Fig. 2 and the and the following text added to the manuscript covers all your concerns.

Lines 367-375: "The total INP concentrations (i.e., the accumulated INP concentration, represented by the sum of each MOUDI stage INP concentration for each sample) at both sites are shown in Fig. 2. Although the INP concentrations measured at both sites were comparable, the exemption was the May 20th sample (Fig. 2a), where higher and statistically significant differences in INP concentrations were measured in the southern site between -19 °C and -22 °C (considering the Agresti and Coull (1998) method to calculate 95 % confidence intervals). Figure 2b also indicates that the INP concentrations from the present study agree well with those reported by Cabrera-Segoviano et al. (2022) for Mexico City and by Chen et al. (2024) for Beijing (between -19 °C and -22 °C), a polluted megacity such as the MCMA.

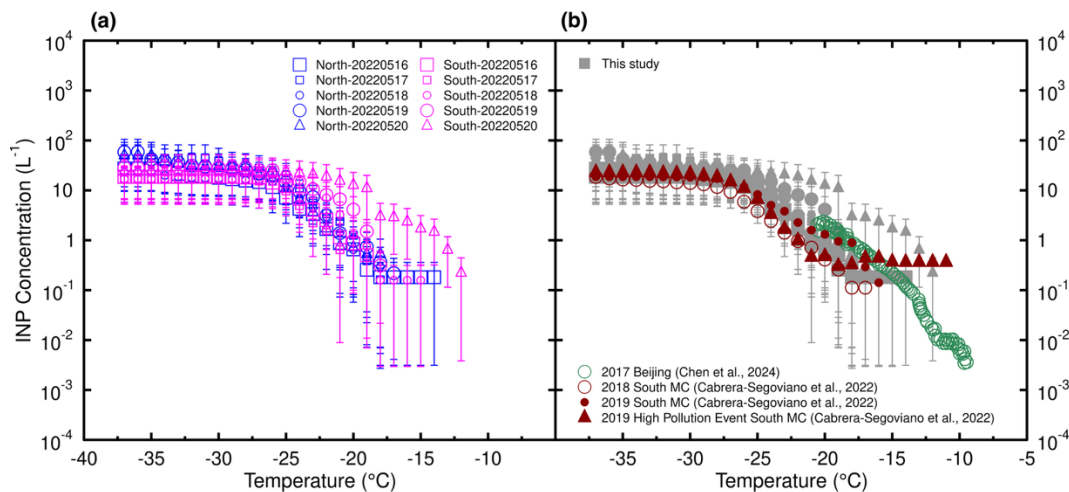


Figure 2. INP concentration as a function of temperature for (a) the measurements performed between May 16th and 20th, 2022 at the northern and southern site of the MCAM, and (b) literature measurements for the MCMA (2018 and 2019), and Beijing (2017) reported by Cabrera-Segoviano et al. (2022) Chen et al. (2024), respectively. The dark red triangles in (b) panel represent the INP concentration values reported for 2019 High Pollution Event at southern Mexico City (Cabrera-Segoviano et al., 2022). The error bars represent the carried experimental uncertainty calculated using the method described in Mason et al. (2015)."

I am additionally aware of the fact that you are also still working with the mean in Fig. 4 and related text. But I would be able to accept that if Fig. 3 shows more clearly all the data you measured, and no misleading mean values, yet.

A/ Regarding Fig. 4 (now presented as Fig. 3), we think that it could be useful to illustrate that the differences between northern and southern site (for bigger particles at -20 °C) can be observed even when using mean values. Demonstrating these differences are clear and this should be studied in more detailed.

Specific comments:

Line 23-24: There is a somewhat misleading sentence early on in your abstract: "However, the aerosol-cloud interaction in megacities, especially in their different microclimates, is poorly understood." You do not really look at aerosol-cloud interactions. That can, by nature, only happen downwind of the emission location. As megacities are large, there may be some influence. But you do not mention at all in your study, how long particles emitted on ground need to reach cloud level height. I opt for deleting this sentence. If you want to keep it, at least change it to: "However, the aerosol-cloud interaction above megacities, especially influenced by their different microclimates, is poorly understood."

A/Thank you for the suggestion. We modified the sentence accordingly.

Lines 23-24: "However, the aerosol-cloud interaction above megacities, especially influenced by their different microclimates, is poorly understood."

Line 37-39: Similar to the last comment, this comment also concerns a sentence in which the importance of considering aerosol-cloud interactions is mentioned, right after you (correctly) state that no strong influence of emissions from the city on INPs was found. Revise the sentence so that it fits the content of the text above.

A/The following text was modified accordingly in the revised manuscript.

Lines 37-39: "This highlights the importance of considering that aerosol-cloud interactions above a megacity may vary, especially when assessing the role of INPs in cloud formation".

Line 573 ff, Chapter 3.3: Repeat explicitly, at a place you find fitting, that measurements were done during a time when vegetation is dry and brown and that results may differ during different times of the year.

A/The following text was modified accordingly in the revised manuscript.

Lines 562-565: "Moreover, the sampling season (dry vegetation season) may be governing the relationship between biological particles and INP concentration reported here, making it necessary to assess INP concentrations at different times of year (i.e., across meteorological seasons)."

Line 600-602 and 603-605: Again, among other factors it depends on your sampling time and volume (and yes, on the atmospheric concentration), if you have a chance to detect these particles or not. So your statement here is not correct per se.

However, when comparing with the concentration range observed already in Petters & Wright (2015), one could argue that with the range of [INP] you can detect, you should have seen bioparticles if they were there. Therefore, I suggest you add at least that "It is still possible that INP contributed by bioparticles are below the detection limit of our setup. However, an overview of INP concentrations observed in the atmosphere by Petters & Wright (2025) indicates that our detection limit of 0.1 1/L is high enough to enable the detection of biological particles if they were present."

A/Thanks a lot for this suggestion. The following text was added in the revised manuscript accordingly.

Lines 554-558: "It is still possible that INP contributed by bioparticles are below the detection limit of our setup because of sampling methods (i.e., differences in cut-off and total sampling time). However, an overview of INP concentrations observed in the atmosphere by Petters & Wright (2015) indicates that our detection limit of 0.1 L-1 is high enough to enable the detection of biological particles if they were present."

Line 639-641: To complete possible reasons, add to the sentence, at the end "or on the dominance of long-range transported INPs, with no or only few additional urban sources".

A/Thank you for this suggestion. The following text was added in the revised manuscript accordingly.

Lines 595-598: "Nevertheless, urban aerosol particles show similar INP concentrations across both sites, suggesting that INP activity does not depend on a specific aerosol type but rather on the bulk complex mixture of aerosol particles or on the dominance of long-range-transported INPs, with no or only a few additional urban sources."

Editorial remarks:

Line 22: Exchange "great" with "major" or "significant", as that may be closer to what you intent to say.

A/Corrected

Line 72: Add "to" after "referred".

A/Corrected

Line 73: Replace "Thanks" with "Owing".

A/Corrected

Line 84: Add "global" before "warming" to be more specific (warming also happens in mid-latitude from winter to summer, ...).

A/Corrected

Line 104: Replace "those of" by a comma. ("... deep convective clouds, which ...")

A/Corrected

Line 146: Replace "along" with "across".

A/Corrected

Line 204-206: How about May 14th and 15th? Some sampling was done on these days, too, as can be seen in Fig. S2. This sentence is misleading, and as the table gives it all well in detail, I suggest to delete this sentence.

A/Corrected

Line 229: Replace "description" by "overview".

A/Corrected

Line 266ff, Chapter 2.2.4: I assumed samples (from shaken filters) were used in both, measurements described in 2.2.3 and here? If that's the case, state this explicitly.

A/Corrected

Line 317: Delete the "and" before "a dry stream".

A/Corrected

Line 454 and related locations in text and supplement: Here, Fig. S2 is mentioned for the first time, after Fig. S3 and S4 were mentioned before. (I had to go back and forth in the supplement to follow your descriptions.) The sequence of appearance of figures in the SI should follow their sequence of appearance in the text. Move the current figure S2 to behind S4, i.e. S3->S2, S4->S3, S2->S4.

A/Thanks a lot for noticing this, but Fig. S2 appears first in the text related to section 2.1 (Lines 198-200): "Meteorological (T, RH, wind direction, wind speed, solar radiation, and precipitation) and criteria pollutants (PM2.5, O3, CO, NOx, and SO2) data were recorded on both sites during the sampling campaign (Tables S1 and S2, and Figs. S1 and S2)."

Line 505: This sentence seems incomplete. Revise. (Maybe you mean: "The fact that CO data is missing for some days on which INP sampling was done may explain the absence of correlations ..." ???)

A/The sentence was corrected

Figure 5: Numbers and asterisks are VERY difficult to read and need to be increased in size.

A/We increased the font size of the figure.

Line 530-532: Where can this rise be seen? Is this visible in any figure? Then relate to that figure. If there is no respective figure, add "(not shown)".

A/Corrected

Line 551: Replace "despised" with "neglected".

A/Corrected

Figure 7: I cannot see any orange markers. Please add or make them larger.

A/Thanks you for pointing this out. We deleted the [PM_2.5] orange markers following other reviewer's suggestion; however, we forgot to remove the text from the figure caption.

Supplement, Figure S6: Some trajectories have rather strange kinks in them, and the trajectories seem to differ from those shown last time. Please make sure this picture is correct!

A/Thanks a lot for checking it. We replace the trajectories in the figure, and we are sure everything is correct.

Literature:

Fletcher, N. H. (1962), The physics of rainclouds, edited, Cambridge University Press.

Petters, M. D., and T. P. Wright (2015), Revisiting ice nucleation from precipitation samples, Geophys. Res. Lett., 42(20), 8758–8766, doi:10.1002/2015gl065733.