

Review of “Dust Transport and Local Anthropogenic Emissions Differently Shape Atmospheric Ice-Nucleating Particles in an Industrial Urban Atmosphere” by Yang et al.

RC1

The manuscript presents one-month measurements of the INP concentration, size distribution and chemical species in the Chinese city of Taiyuan. While for days influenced by dust transport an enhanced INP concentration and inas density was observed, pollution episodes did not influence the aerosols' freezing ability substantially. As none of the chemical components correlated with the INP concentration during non-dust days, they concluded that the INP population in these times is composed of an interplay of particles rather than one single source.

Overall, the manuscript is well written and suits the scope of the journal. However, there are some points, listed below, that need clarification and would further improve the quality of the paper. After addressing these comments, I would recommend the manuscript for publication.

Response: We thank the reviewer for the careful reading of our manuscript and for the positive overall assessment. We appreciate the reviewer's concise summary of the main findings and the recognition that the manuscript is generally well written and within the scope of the journal. We have carefully considered all of the comments below and revised the manuscript accordingly. Detailed responses are provided point by point.

General comments

In L.172 you state that the surface area A , used to calculate n_s , is obtained from the SMPS measurements. According to L.111 the SMPS data cover the diameter range from 3 nm–453 nm. With that, you leave out the particles larger than 0.5 μm in size, which are thought to be the most active in ice formation and many parameterizations are based on that. Even though the number of particles larger than 0.5 μm in size might be low, the total surface area is strongly affected. I am aware that the conversion from the optical to the aerodynamic diameter may introduce some uncertainty, however, not including these particles in the surface area will increase the uncertainty of n_s . I would suggest calculating n_s with the combined size distribution of the SMPS and OPC and provide a detailed discussion on the uncertainty of the surface area and with that n_s . Furthermore, the discussion around Fig. 2 and the comparison to literature data and parameterizations should be revised.

Response: We thank the reviewer for this important comment. In the actual n_s calculation, the total particle surface area A was derived from the combined particle size distributions measured by the SMPS and OPC, rather than from SMPS data alone. Thus, the contribution of particles larger than 0.5 μm was included in the calculation. The sentence stating that “ A was calculated based on concurrent SMPS data” was inaccurate and has been revised accordingly. We have revised the manuscript to clarify this point explicitly and to better describe how the merged SMPS–OPC size distribution was used to estimate A for n_s calculation. We have also clarified the uncertainty

associated with diameter conversion and surface-area estimation, and revised the relevant text accordingly.

Changes in manuscript:

(1) Section 2.2.1: “The OPC data were combined with the SMPS measurements to construct a merged particle size distribution (SMPS: 3–453 nm; OPC: 0.5–20 μm), which was further used to estimate the total particle surface area for n_s calculation.”

(2) Section 2.2.3: “...and A was calculated from the combined particle size distributions measured by the SMPS and OPC.”

(3) Section 3.3: “The n_s values discussed here were calculated from the merged SMPS–OPC size distribution, thereby accounting for contributions from coarse particles in the surface-area estimate.”

Minor comments

- L.92: Please add the altitude of the measurement site

Response: We have added the approximate ground elevation of the sampling site in the revised manuscript.

Changes in manuscript:

Section 2.1: “The sampling site (37.88°N, 112.55°E; approximately 800 m a.s.l.) is located on the rooftop (~20 m above ground level) of a four-story building at the Taiyuan Municipal Ecology and Environment Bureau.”

- L.102: What is a two-channel sampler? Did you sample 2 filters in parallel with that? And what is your sampling cut-off? Please provide more details on that.

Response: Here, “two-channel sampler” refers to a sampler equipped with two independent parallel filter channels, allowing two 47 mm filters to be mounted and sampled simultaneously. In this study, the sampler was operated without a cyclone or other size-selective inlet; therefore, no specific aerodynamic cut-off was applied, and the collected aerosol can be regarded as approximately total suspended particles (TSP). We have revised the manuscript to clarify these details.

Changes in manuscript:

Section 2.1: “In addition, INP filter samples were collected using a two-channel sampler equipped with two parallel filter channels from 4 December 2023 to 5 January 2024. Polycarbonate filters (47 mm, Nuclepore Track-Etch Membrane, 0.2 μm pore size, Whatman) were used for sampling. The sampler was operated without a size-selective inlet.”

- L.121: Why did you convert everything to the mobility diameter? In the manuscript you don’t pick it up again, so I think you should discuss the conversion of the mobility diameter to the aerodynamic diameter and give an uncertainty for that.

Response: The size distributions were expressed in mobility diameter because the particle surface area used for n_s calculation needs to be calculated on the same diameter basis. The SMPS data are given in mobility diameter, whereas the OPC measures optical-equivalent diameter for larger particles. Therefore, the OPC size distribution was converted to mobility diameter before being merged with the SMPS data for surface-area calculation.

The main uncertainty in this conversion comes from the assumptions used in the conversion. In the revised manuscript, we now state that an effective particle density of 1.5 g cm^{-3} and a shape factor of 1 were assumed. Chen et al. (2021) evaluated the effect of particle density and shape factor on n_s calculation and showed that changing the particle density from 1.5 to 2.6 g cm^{-3} and the dynamic shape factor from 1.1 to 1.4 can lead to a factor of 1.07 – 2.36 difference in n_s . This result indicates the possible magnitude of uncertainty caused by diameter conversion. For our dataset, an exact uncertainty cannot be assigned because particle refractive index, morphology, and sample-specific effective density were not independently measured. We have clarified these assumptions and the related uncertainty in the revised manuscript.

Changes in manuscript:

Section 2.2.1: “To merge the OPC data with the SMPS measurements and calculate particle surface area on the same diameter basis, the OPC size distribution was converted to mobility diameter (d_m). Estimating d_a from optical-equivalent diameter typically requires knowledge of particle complex refractive index and morphological parameters (e.g., dynamic shape factor) (Peters et al., 2006). Moreover, when converting to d_m , we assumed an effective particle density of 1.5 g cm^{-3} and a shape factor of unity. Previous sensitivity calculations showed that changing the particle density from 1.5 to 2.6 g cm^{-3} and the dynamic shape factor from 1.1 to 1.4 can lead to a factor of 1.07 – 2.36 difference in n_s (Chen et al., 2021). This suggests that diameter conversion may introduce an uncertainty of approximately a factor of 2 in n_s , particularly in the coarse-mode size range (Huang et al., 2021). A more exact uncertainty could not be assigned because particle refractive index, morphology, and particle-specific effective density were not independently measured.”

- L.122: Here you should give the value of the uncertainty. I think this is an important addition also for the discussion of n_s , which should include the larger particles (see my comment in the general comments section)

Response: The uncertainty of n_s cannot be represented by a single well-constrained numerical value from the present measurements. This is because n_s depends on both N_{INP} and the particle surface area concentration used for normalization. The uncertainty in N_{INP} mainly arises from the statistical limitation of the droplet-freezing experiment, as described in Sect. 2.2.3. The uncertainty in the particle surface area concentration is mainly associated with the merged SMPS–OPC size distribution, especially the conversion of OPC optical-equivalent diameters in the coarse-mode range. This conversion requires assumptions regarding particle density and shape factor, while particle-specific refractive index, morphology, and effective density were not independently measured. Therefore, we did not assign an unsupported single numerical uncertainty to n_s , but clarified the main uncertainty sources in the revised manuscript. The contribution of coarse-mode particles was included in the SMPS–OPC merged particle surface area used for n_s calculation, as also clarified in the response to the general comment above.

Changes in manuscript:

Section 2.2.3: “For each aerosol sample, the parameters ($f_{\text{ice}}(T)$, $K(T)$, $N_{\text{INP}}(T)$, $n_s(T)$, and the confidence intervals for $N_{\text{INP}}(T)$) were calculated individually. The $f_{\text{ice}}(T)$ was

derived from cold-stage measurements, V_{air} was obtained from recorded sampling volumes, and A was calculated from the standard-condition combined particle size distributions measured by the SMPS and OPC. The uncertainty in n_s arises from both the statistical uncertainty in N_{INP} and the uncertainty in the particle surface area concentration used for normalization. The latter is mainly associated with the SMPS–OPC merging and diameter conversion in the coarse-mode range, as discussed in Section 2.2.1.”

- L.153: Were also the aerosol data converted to standard conditions?

Response: In the original calculation, the INP filter-sampling volume from the two-channel sampler used to derive N_{INP} had been converted to standard conditions (0 °C, 1013 hPa). Following the reviewer’s comment, we have also converted the particle number size distributions to the same standard-condition air-volume basis using concurrent temperature and pressure data. The particle number and surface area concentrations derived from the size distributions were then recalculated and used to update the corresponding n_s values and related figure data.

This correction caused only minor quantitative changes. For example, the daily correction factor for the particle surface area concentration ranged from 0.981 to 1.142, with a mean value of 1.079 during the INP observation period. The recalculated n_s values were slightly lower on average, but the main patterns and conclusions were unchanged.

Changes in manuscript:

Section 2.2.3: “For $n_s(T)$ calculation, the merged SMPS–OPC particle number size distributions were converted to the same standard-condition air-volume basis as N_{INP} (0 °C, 1013 hPa) using concurrent temperature and pressure data. The particle surface area was then estimated from the converted size distributions expressed in mobility diameter.”

- L.178: Please specify why you chose 850 m a.s.l.; is it the height of the station?

Response: The arrival height of 850 m a.s.l. was chosen to approximately represent the actual sampling inlet height, considering that the ground elevation of the sampling site is about 800 m a.s.l. and the sampling inlet was located on the rooftop at about 20 m above ground level. We have clarified this in the revised manuscript.

Changes in manuscript:

Section 2.2.4: “Trajectories were calculated with a duration of 72 h and an interval of 6 h between each trajectory, arriving at an altitude of 850 m above sea level (a.s.l.), which approximately represents the actual sampling inlet height, considering a ground elevation of about 800 m a.s.l. and a rooftop sampling height of about 20 m above ground level.”

- L.206: You discuss here the INP concentration at a temperature of -15 °C. However, looking into Fig. 1a, it is not clear which of the temperature ranges you mean (-12.5 °C–-15 °C or -15 °C to 17.5 °C). Please clarify it in the text.

Response: Here, the INP concentration at -15 °C refers to the cumulative $N_{\text{INP}}(T)$ evaluated at $T = -15$ °C, i.e., the total INP concentration active at and above this temperature, rather than to a specific temperature interval shown in Fig. 1a. We have

clarified this in the revised manuscript.

Changes in manuscript:

Section 3.1: “At and above $-15\text{ }^{\circ}\text{C}$, the cumulative $N_{\text{INP}}(T)$, representing the total INP concentration active at and above this temperature, ranged from 10^{-2} to 10^1 L^{-1} with a campaign mean of 1.75 L^{-1} .”

- L.207: Do you refer to the n_s values seen in Fig. 3? Either add the reference to the figure or add the n_s values in Fig. 1.

Response: The n_s values referred to here are those shown later in Fig. 3. To avoid overcrowding Fig. 1, we have added an explicit reference to Fig. 3 in the revised text rather than adding n_s values to Fig. 1.

Changes in manuscript:

Section 3.1: “Correspondingly, the n_s values at and above $-15\text{ }^{\circ}\text{C}$ (see Fig. 3) spanned 10^5 to 10^7 m^{-2} , with a campaign mean of $3.42 \times 10^6\text{ m}^{-2}$.”

- Fig. 1a: The color of the bars of the INP concentration seems to change when you mark the special periods such as the dust transport. For more clarity, please make sure that the shaded areas are in the back and do not change the color the other markers.

Response: We have revised Fig. 1a by replacing the original shading with dashed boundary lines and arrow annotations to indicate the special periods. This avoids interference with the original colors of the INP bars and improves the clarity of the figure.

Changes in manuscript:

(1) Section 3.1: “Several distinct events can be identified throughout the campaign, as indicated in Fig. 1 by dashed boundary lines and colored arrows. The first event (5–9 December, marked by the orange arrow in Fig. 1a) was characterized by a sharp increase in coarse-mode (here defined as particles with aerodynamic diameter $>1\text{ }\mu\text{m}$, $N_{>1\mu\text{m}}$) particle concentrations (Figs. 1b, S1b, and S2).”

(2) Section 3.1: “Other episodes (13–14, 18, and 28–30 December, marked by the gray arrows in Fig. 1a) were characterized by simultaneous increases in sulfate, nitrate, ammonium, and organic carbon, accompanied by elevated both $\text{PM}_{2.5}$ and PM_{10} mass (Fig. 1c and Fig. 1d).”

(3) Section 3.3: “During the desert dust event (5–9 December; marked by the orange arrow and dashed boundary lines in Fig. 1a), both N_{INP} and n_s increased markedly.”

(4) Figure 1 caption added: “Special periods are indicated by dashed boundary lines across all panels and colored arrows at the top of panel (a): the orange arrow denotes the desert dust period, the gray arrows denote pollution periods, and the red arrow denotes the fireworks event.”

- Fig. 1a: The gray triangles of the DeMott et al., 2015 parameterization should be removed here. You can add them to Fig. 2, where you anyways discuss some INP parameterizations.

Response: We understand the reviewer’s concern regarding figure clarity. However, we prefer to retain the DeMott et al. (2015) parameterization in Fig. 1a, because its purpose here is not primarily to provide a general parameterization comparison, but to show the day-to-day difference between the D15-predicted INP concentrations and the observed

values during different periods of the campaign. Since the D15 values were calculated using the corresponding daily coarse-particle number concentrations, placing them in the time-series panel allows a direct temporal comparison with the observations. By contrast, Fig. 2 is mainly intended for temperature-dependent comparison with previous studies and reference parameterizations, and moving D15 there would weaken its value as a temporal diagnostic. To improve readability, we have revised Fig. 1a and clarified the role of D15 in both the figure caption and the main text.

Changes in manuscript:

(1) Figure 1a has been revised to improve clarity while retaining the D15(−20 °C) time-series comparison.

(2) Figure 1 caption: “In panel (a), gray triangles (D15(−20 °C)) represent values calculated using the NINP parameterization scheme proposed by DeMott et al. (2015), shown here to compare the day-to-day temporal variation between parameterized and observed INP concentrations.”

(3) Section 3.3: “Figure 1a also compares the measurements with the DeMott et al. (2015) parameterization (D15), allowing a direct day-to-day comparison between the parameterized and observed INP concentrations during different periods of the campaign.”

- L.236: Cu and Sr are not shown in Fig. 1c or 1d.

Response: These species were measured and showed concurrent enhancements during the fireworks event, but they were not retained in the final version of Fig. 1 in order to avoid overloading the panel. The text was not updated accordingly, which caused the inconsistency. We have now revised the text to match the current figure.

Changes in manuscript:

Section 3.1: “Around 2 January 2024 (marked by the red arrow in Fig. 1a), a short-lived pollution event occurred, accompanied by sharp increases in PM mass and concurrent peaks in K, OC, and EC (Figs. 1c and 1d), which are consistent with firework emissions during New Year celebrations.”

- Fig. 2: Your freezing spectra of the dust episode have a very interesting profile, which is very characteristic of biological aerosol particles and not in the first place of dust. Hu et al., 2023 observed a similar INP concentration in Beijing in spring during dust transport. They performed a heat treatment on the samples and concluded that there might be some mixing of the dust aerosol with more heat sensitive, potentially biological, particles. Did you also perform a heat treatment to your samples? I think the discussion you provide in L.310 ff should be provided already here, with reference to the above-mentioned paper.

Response: We did not perform heat-treatment experiments on these samples and therefore cannot directly evaluate the contribution of heat-sensitive biological components in this study. However, the revised manuscript now acknowledges this possibility earlier in the discussion associated with Fig. 2. Previous heat-treatment results during East Asian dust events showed that heat-sensitive INPs made substantial contributions at relatively warm temperatures (Chen et al., 2021), and Hu et al. (2023) reported similar behavior during spring dust transport in Beijing. We now state that the enhanced warm-temperature activity during the Taiyuan dust episode may reflect the

combined influence of transported mineral dust and potentially co-transported heat-sensitive components, although such a contribution can only be inferred and cannot be directly assessed in the absence of heat-treatment analysis in this study.

Changes in manuscript:

Section 3.2: “However, the enhanced warm-temperature INP activity during this episode may not be fully attributable to mineral dust alone. During East Asian dust events, Chen et al. (2021) showed that heat-sensitive INPs made substantial contributions at relatively warm temperatures. Similarly, Hu et al. (2023) reported enhanced warm-temperature INP activity during spring dust transport in Beijing and suggested, based on heat-treatment analysis, that transported dust particles may be mixed with heat-sensitive, potentially biological components. Together, these studies support the possibility of similar heat-sensitive contributions during the Taiyuan dust episode. In the present study, however, no heat-treatment analysis was performed; therefore, such a contribution can only be inferred and cannot be directly assessed.”

- L.274: should be orange instead of light gray

Response: The corresponding text has been revised from “light gray” to “orange”.

- L.278 ff: DeMott et al., 2015 is a parameterization made for desert dust aerosol. While for the dust episode it is a valid choice, it is expected that for the remaining measurements it might not provide reasonable results. Please specify this in the text.

Response: We agree that the DeMott et al. (2015) parameterization was developed for desert dust aerosol and is therefore most appropriate for interpreting the dust episode in this study. For the non-desert-dust periods, we do not use D15 as a general predictor, but rather as a dust-based reference to show how the observed INP concentrations deviate from desert-dust-dominated behavior. We have clarified this point in the revised manuscript and now explicitly state that the interpretation of D15 during non-desert-dust periods should be made with caution.

Changes in manuscript:

Section 3.3: “Figure 1a also compares the measurements with the DeMott et al. (2015) parameterization (D15), which links INP concentrations to the number concentration of particles larger than 0.5 μm and was originally developed for desert dust aerosol. Accordingly, it is most appropriate for the dust episode in this study, whereas for the remaining periods it is shown mainly as a dust-based reference and should be interpreted with caution.”

- L.340: Did you also check the same correlations for other temperatures?

Response: We have additionally examined the correlations between the daily mean PMF-resolved source contributions and N_{INP} at -10 , -12.5 , -15 , and -17.5 $^{\circ}\text{C}$, in addition to -20 $^{\circ}\text{C}$. Values indicating complete freezing before the corresponding temperature were excluded from the correlation analysis. As shown in the newly added Table S2, none of the PMF-resolved factors showed statistically significant correlations with N_{INP} at any examined temperature ($p > 0.05$). Therefore, the main interpretation is not sensitive to the selected temperature.

Changes in manuscript:

Section 3.4: “Similar analyses were also conducted at -10 , -12.5 , -15 , and -17.5 $^{\circ}\text{C}$ (Table S2). No statistically significant correlations were found between N_{INP} and any

PMF-resolved factor at the examined temperatures ($p > 0.05$), indicating that this result was not specific to the choice of $-20\text{ }^{\circ}\text{C}$.”

- Conclusions: When you discuss your findings during the dust transport episode, you should mention that potential aging processes do not suppress the INP efficiency. Next to that, you should mention again the potential mixing with biogenic particles. Since they are very ice active, it could be that you see in your samples primarily the biogenic particles freezing rather than the dust particles.

Response: In the revised manuscript, we now state that comparison with previously reported dust parameterizations indicates that transported desert dust retained strong ice-nucleating activity during the dust transport episode. We also reiterate that the enhanced warm-temperature activity may reflect not only transported mineral dust, but also the possible contribution of co-transported biological components. However, because no heat-treatment analysis was performed in this study, this remains a plausible interpretation rather than a direct confirmation.

Changes in manuscript:

Conclusions: “Comparison with previously reported dust parameterizations indicates that the transported desert dust retained strong ice-nucleating activity during the dust transport episode. The enhanced warm-temperature activity may also reflect the possible contribution of co-transported biological components, although this cannot be directly verified in the absence of heat-treatment analysis.”

References

Hu, Y., Tian, P., Huang, M., Bi, K., Schneider, J., Umo, N. S., Ullmerich, N., Höhler, K., Jing, X., Xue, H., Ding, D., Liu, Y., Leisner, T., and Möhler, O.: Characteristics of ice-nucleating particles in Beijing during spring: A comparison study of measurements between the suburban and a nearby mountain area, *Atmospheric Environment*, 293, 119451, <https://doi.org/10.1016/j.atmosenv.2022.119451>, 2023.

Chen, J., Wu, Z., Chen, J., Reicher, N., Fang, X., Rudich, Y., and Hu, M.: Size-resolved atmospheric ice-nucleating particles during East Asian dust events, *Atmospheric Chemistry and Physics*, 21, 3491–3506, <https://doi.org/10.5194/acp-21-3491-2021>, 2021.

RC2

This study assessed the ice nucleating particle (INP) concentrations and efficiencies in an urban site in China. The authors were able to identify a clear mineral dust event as well as other high pollution events during their sampling period. Additionally, most of the sampling days were assigned as a “non-dust period” considering aerosol composition, particle size distributions, and air masses history. Although INPs are poorly constrained and studied in urban sites, the manuscript requires deep corrections. Also, I think this manuscript better fits into the “Measurement report” category.

Response: We thank the reviewer for the careful reading of our manuscript and for recognizing the value of the Taiyuan INP dataset. We appreciate the reviewer’s constructive comments and have carefully revised the manuscript accordingly. Detailed point-by-point responses are provided below.

Major comments:

1. The authors made a distinction between a “dust event” and a “non-dust period”. Although the INP concentration and ns values are quite different between both periods, I am not fully convinced that the “non-dust period” is really a period without the influence of mineral dust particles. As shown in the back-trajectories, during the “non-dust period” the airmasses crossed northern China and southern Mongolia deserts, including the Gobi Desert. Also, the elemental analysis is not a good way to confirm the absence of mineral dust particles during the “non-dust period” as aluminum (Al) and silicon (Si) were not included. This is a key point to be addressed as most of the drawn conclusions took this apparent distinction into account.

Response: We agree that the term “non-dust period” in the original manuscript was not sufficiently precise, because it may imply a complete absence of mineral dust influence. Our intention was only to distinguish the clearly identified desert dust intrusion event (5–9 December) from the remaining observation periods. We have therefore revised the terminology throughout the manuscript to “non-desert-dust periods” (or equivalent cautious wording, depending on context). We now clarify that the desert dust event was identified based on multiple lines of evidence, including backward trajectories, coarse-mode particle enhancement, and elevated PM₁₀. Outside this event, we do not claim that mineral dust was absent, but only that no similarly clear desert dust intrusion signature was identified. The relevant interpretations have been revised accordingly.

Changes in manuscript:

Abstract: “with the remaining observation periods without clear desert dust intrusion”

Section 3.1: “which was approximately 2.7 times higher than the average concentration during the remaining observation period”

“The remaining observation periods are not assumed to be free of mineral dust influence; rather, no similarly clear desert dust intrusion signature was identified outside this event.”

Section 3.2: “Taken together, these features indicate that episodic desert dust transport was associated with the most prominent INP enhancements during this campaign, whereas the remaining variability during the non-desert-dust periods likely reflects a more complex mixture of sources and particle properties that cannot be uniquely resolved by the present dataset.”

Section 3.4, title: The section has been revised from “Anthropogenic particle pollution affecting INP concentrations during non-dust periods” to “Anthropogenic particle pollution and INP variability during non-desert-dust periods” to avoid implying a direct influence in the section heading.

Section 3.4: “During the non-desert-dust periods, this factor is interpreted as mainly reflecting local dust-associated particles, such as road dust, fugitive dust from industrial and construction activities, and resuspended soil. This dust-related factor showed no significant correlation with N_{INP} abundance (Table 1).”

“These results suggest that, during non-desert-dust periods of this campaign, N_{INP} variability was not clearly controlled by any of the major PMF-resolved source

categories. This is consistent with previous observations at urban sites in different regions, where INPs remain scarce and episodic relative to total aerosol loading, and where their variability is often not well explained by the major anthropogenic aerosol components.”

“The results provide observational constraints on wintertime INP abundance and variability in an industrial urban atmosphere.”

“A key finding of this winter campaign is that the strongest INP enhancements in Taiyuan were associated with episodic natural desert dust transport.”

“Overall, the observations from this winter campaign indicate that the strongest INP enhancements in Taiyuan were associated with long-range desert dust transport, whereas the remaining INP variability during the non-desert-dust periods reflected more complex influences that were not resolved by the major PM_{2.5} source factors considered here. These results provide observational constraints for understanding wintertime urban INPs and for improving their representation in chemical transport and climate models.”

2. The size range or cut-off size of the INP samples is not provided. This is very important as it is well-known that particle size is a key variable when assessing their ice-nucleating abilities. I only found the following information: “using a two-channel sampler without a cyclone”. Does it mean that the samples correspond to Total suspended particles (TSP)? Did I miss something?

Response: The INP filter samples in this study were collected without a cyclone, impactor, or other size-selective inlet. Therefore, no defined aerodynamic cut-off was applied during sampling, and the collected aerosol can be regarded as approximately total suspended particles (TSP). We have clarified it explicitly in the revised manuscript.

Changes in manuscript:

Section 2.1: “In addition, INP filter samples were collected using a two-channel sampler equipped with two parallel filter channels from 4 December 2023 to 5 January 2024. Polycarbonate filters (47 mm, Nuclepore Track-Etch Membrane, 0.2 μm pore size, Whatman) were used for sampling. The sampler was operated without a size-selective inlet.”

3. The PMF analysis was performed on PM_{2.5} particles; however, as mentioned above in point #2, based on the provided information, it seems that the INP samples correspond to TSP. If this is the case, the performed correlations are meaningless.

Response: We agree that this comparison has a clear limitation, because the PMF analysis was based on PM_{2.5} chemical composition, whereas the INP filter samples are more appropriately regarded as approximately representative of TSP. Thus, the PMF-resolved factors do not capture the full particle size range contributing to the filter-based INP measurements. Our intention, however, was not to use PMF for direct source attribution of INPs, but to examine whether the dominant PM_{2.5} source factors covaried with the observed N_{INP} variability during the non-desert-dust periods. We have clarified this limitation in the revised manuscript and revised the discussion accordingly, so that the PMF–N_{INP} relationships are presented more cautiously as covariation with major

PM_{2.5} source factors rather than direct source attribution for INPs.

Changes in manuscript:

Section 3.4: “To further examine whether the dominant PM_{2.5} source factors were associated with N_{INP} variability during the non-desert-dust periods, PMF analysis was applied to the PM_{2.5} chemical composition dataset, resolving five major factors...”

“To evaluate whether any of these major PM_{2.5} sources covaried with INP, Pearson correlations...”

“All correlations were weak ($|r| < 0.3$) and statistically insignificant ($p > 0.05$), indicating that the dominant PM_{2.5} source factors did not show clear covariation with INP abundance.”

4. I was surprised that meteorology was not used at all. The authors argue in several parts that the INPs were from local sources during the “non-dust period”. It would have been nice to check the wind speed and wind direction to corroborate this.

Response: We thank the reviewer for this helpful comment. We examined the wind data and added wind rose plots for the identified dust event and the non-desert-dust periods to the Supplement (Fig. S6). These plots show that both periods were dominated by northerly flow, while the dust-event period exhibited a more concentrated northerly flow pattern. We have added a brief statement in the main text to refer to this result as supplementary meteorological support for the event classification. At the same time, we avoid overinterpreting the wind data as definitive source-direction attribution for INPs.

Changes in manuscript:

Section 3.1: “Wind rose analysis (Fig. S6) shows that both the dust-event and non-desert-dust periods were dominated by northerly flow, whereas the dust-event period exhibited a more concentrated northerly flow pattern. This serves as supplementary meteorological support for the event classification.”

Supplement: Wind rose plots for the identified dust event and the non-desert-dust periods have been added to provide meteorological context for the event classification.

5. Figures S4 and S5 are not mentioned/discussed in the main text. These figures contain important information that deserves to be deeply discussed.

Response: In the revised manuscript, we have added explicit references to these figures in Section 3.4 and briefly discussed how they support the PMF factor assignments.

Changes in manuscript:

Section 3.4: An explicit reference to Figs. S4 and S5 has been added in Section 3.4 to show that the PMF factor profiles and inter-species correlation patterns provide supplementary support for the factor assignments.

“The corresponding factor profiles (Fig. S4) are consistent with the assigned source categories, including crustal tracers in the dust-related factor and Cu–Ba–K enrichment in the fireworks factor. The inter-species correlation patterns shown in Fig. S5 also support these factor assignments.”

6. “All samples in Taiyuan activated before $-15\text{ }^{\circ}\text{C}$, with the most active samples

initiating freezing near -5°C ” and “It is also noteworthy that at relatively warm temperatures (approximately $T > -12^{\circ}\text{C}$), ns values of the ambient desert dust samples substantially exceed the parameterizations for pure K-feldspar”. If this is true, this corresponds to extremely efficient INPs; however, a deep discussion on the high efficiency of these particles is not provided. Is this because the samples were TSP? Is this coming from an instrumental bias? Were these samples enriched in biological particles?

Response: The revised manuscript expands the discussion to address the three possibilities raised by the reviewer. First, because the INP samples were collected without a size-selective inlet, the filter samples can be regarded as approximately TSP and may have retained coarse particles, including potentially biologically derived material. However, this sampling configuration was the same for all samples and therefore cannot by itself explain why the strongest warm-temperature activity was observed specifically in the dust-affected samples. Second, instrumental bias is unlikely to be the main explanation, because the warm-temperature enhancement was event-specific rather than a general feature of all samples measured with the same cold-stage-based approach. Third, enrichment of heat-sensitive, potentially biological components remains a plausible explanation. Previous heat-treatment results during East Asian dust events showed that heat-sensitive INPs contributed substantially at relatively warm temperatures (Chen et al., 2021), and Hu et al. (2023) reported similar behavior during spring dust transport in Beijing. We have therefore revised the discussion to state that this elevated warm-temperature activity may reflect the combined influence of transported mineral dust and potentially co-transported heat-sensitive components, although such a contribution can only be inferred and cannot be directly assessed in the absence of heat-treatment analysis in this study.

Section 3.2: “However, the enhanced warm-temperature INP activity during this episode may not be fully attributable to mineral dust alone. During East Asian dust events, Chen et al. (2021) showed that heat-sensitive INPs made substantial contributions at relatively warm temperatures. Similarly, Hu et al. (2023) reported enhanced warm-temperature INP activity during spring dust transport in Beijing and suggested, based on heat-treatment analysis, that transported dust particles may be mixed with heat-sensitive, potentially biological components. Together, these studies support the possibility of similar heat-sensitive contributions during the Taiyuan dust episode. In the present study, however, no heat-treatment analysis was performed; therefore, such a contribution can only be inferred and cannot be directly assessed.”

Section 3.3: “The TSP-like sampling configuration may have facilitated retention of such coarse ice-active components, but it cannot by itself explain the selective enhancement observed during the dust event.”

7. Why the PMF analysis was correlated with the INP concentration at -20°C only. I suggest testing this comparison at -10°C and -15°C as this refers to highly efficient INPs.

Response: Following the reviewer’s suggestion, we extended the Pearson correlation analysis to N_{INP} at -10 , -12.5 , -15 , and -17.5°C , in addition to -20°C . Values beyond

the quantifiable range due to complete freezing before the corresponding temperature were excluded from the analysis. No statistically significant correlations were found between N_{INP} and any PMF-resolved factor at the examined temperatures ($p > 0.05$). These additional results are now provided in Table S2, and the main text has been revised accordingly.

Changes in manuscript:

Section 3.4: “Similar analyses were also conducted at -10 , -12.5 , -15 , and -17.5 °C (Table S2). No statistically significant correlations were found between N_{INP} and any PMF-resolved factor at the examined temperatures ($p > 0.05$), indicating that this result was not specific to the choice of -20 °C.”

Supplement, Table S2: Pearson correlation coefficients between daily mean PMF-resolved source contributions and N_{INP} at different temperatures during non-desert-dust periods were added.

Minor comments:

Lines 19-20: “compared with periods without natural dust influence”. I don’t think the authors can completely rule out the presence of mineral dust particles in the “non-dust period”.

Response: We agree that the original wording in the Abstract was too strong, because it could imply a complete absence of mineral dust influence outside the identified dust event. We have therefore revised this phrase to a more cautious expression referring to the remaining observation periods without clear desert-dust intrusion.

Changes in manuscript:

Abstract: “compared with the remaining observation periods without clear desert-dust intrusion”

Lines 26-27: “Observed N_{INP} variability is likely governed by the interplay of episodic coarse-mode inputs and atmospheric processing rather than a single dominant source.” I am not sure the authors can really say this as a direct correlation between INP concentration and coarse particle concentration was not provided.

Response: We have revised this sentence in the Abstract to a more observation-based summary limited to this winter campaign.

Changes in manuscript:

Abstract: “During this winter campaign, N_{INP} variability in Taiyuan was characterized by strong enhancement during the identified desert dust event and weak associations with the major $\text{PM}_{2.5}$ source factors during the remaining observation periods.”

Line 178: What was the main motivation to run the back-trajectories at 859 m a.s.l.?

Response: The arrival height of 850 m a.s.l. was chosen to approximately represent the actual sampling inlet height, considering that the ground elevation of the sampling site is about 800 m a.s.l. and the sampling inlet was located on the rooftop at about 20 m above ground level. We have clarified this in the revised manuscript.

Section 2.2.4: “Trajectories were calculated with a duration of 72 h and an interval of 6 h between each trajectory, arriving at an altitude of 850 m above sea level (a.s.l.),

which approximately represents the actual sampling inlet height, considering a ground elevation of about 800 m a.s.l. and a rooftop sampling height of about 20 m above ground level.”

Lines 249-250: “while it is comparable to observations from Sisal, Mexico (Ladino et al., 2016), and New Delhi, India (Wagh et al., 2021) between -15°C to -20°C ”. I disagree with this statement. It would have been better to add to Figure 2 in-situ data

Response: The New Delhi dataset has been removed from Fig. 2 because it was obtained in the deposition mode. We also removed the direct “comparable to Sisal and New Delhi” statement from Section 3.2. To provide a more relevant comparison for the present winter campaign, we added wintertime urban INP observations from Beijing (Chen et al., 2018) and revised the Tokyo comparison to use wintertime data from Tobo et al. (2020). The associated text has been softened to present these datasets as observational context rather than direct source attribution.

Changes in manuscript:

Section 3.2: Section 3.2 and Fig. 2 were revised by removing the New Delhi comparison, adding wintertime Beijing observations from Chen et al. (2018), using wintertime Tokyo observations from Tobo et al. (2020), and softening the comparison with previous studies.

Lines 252-253: “This implies that the INPs observed in Taiyuan may also be influenced by anthropogenic dust particles.” No information is provided to support this.

Response: We thank the reviewer for this important comment. Our intention was not to infer anthropogenic dust influence in Taiyuan directly from Chen et al. (2024), but rather to note that anthropogenic dust has been reported as an important urban INP source in some environments. To avoid this ambiguity, we have removed the above sentence and revised the surrounding discussion. The revised text now states more cautiously that Chen et al. (2024) and the present study together suggest that the relative importance of anthropogenic dust and transported desert dust may vary among urban environments and observation periods. In the present winter campaign in Taiyuan, the strongest INP enhancements were associated with the identified desert dust event, whereas the remaining variability during non-desert-dust periods could not be uniquely attributed to anthropogenic dust based on the present dataset.

Changes in manuscript:

Section 3.2: The sentence “This implies that the INPs observed in Taiyuan may also be influenced by anthropogenic dust particles” has been removed.

Line 256: “they are broadly comparable with those reported in other relevant studies.” I disagree with this statement.

Response: We have removed this wording and retained only the more specific comparisons presented in the surrounding text.

Section 3.2: The sentence “Although the N_{INP} levels in Taiyuan are relatively high compared with other urban observations, they are broadly comparable with those reported in other relevant studies” has been removed.

Lines 257-258: I don't think that it is a good idea to add the data from Petters and Wright (2015). I suggest removing it from Figure 2.

Response: We have revised the text to clarify that this range is retained only as a broad background reference for atmospheric INP abundance, rather than as evidence that the Taiyuan measurements are directly comparable to precipitation samples. The corresponding discussion has been softened accordingly.

Changes in manuscript:

Section 3.2: "Additionally, the gray shaded area denotes the N_{INP} range derived from precipitation samples summarized by Petters and Wright (2015), which is used here as a broad reference for background atmospheric INP abundance. At temperatures warmer than $-15\text{ }^{\circ}\text{C}$, the Taiyuan N_{INP} values fall mostly within this range during the non-desert-dust periods."

Lines 261-262: "suggesting additional contributions from ice-active components or enhanced influence of transported desert dust during certain periods." This is contradictory as this data corresponds to the "non-dust period".

Response: We agree that the original wording was contradictory, because this part of the discussion referred to the non-desert-dust periods and therefore should not imply enhanced influence of transported desert dust. Our intention was only to indicate that, at colder temperatures, the measured N_{INP} values exceeded the reference range, suggesting the presence of additional ice-active components active at lower temperatures. We have revised the sentence accordingly.

Changes in manuscript:

Section 3.2: "At colder temperatures, however, the measured N_{INP} values exceed this range substantially, suggesting contributions from additional ice-active components active at lower temperatures."

Lines 283-284: "particles larger than $0.5\text{ }\mu\text{m}$ in Taiyuan are likely dominated by local surface emissions from the surrounding Loess Plateau and fugitive dust from intensive industrial activities". No evidence is provided.

Response: Our intention was only to suggest that coarse particles present during non-desert-dust periods may differ in source and ice-nucleating efficiency from the transported desert dust represented by the D15 parameterization. We have therefore revised this sentence to a more cautious wording.

Changes in manuscript:

Section 3.3: "This discrepancy suggests that coarse-particle abundance alone may not fully constrain INP variability in Taiyuan. Particles larger than $0.5\text{ }\mu\text{m}$ in Taiyuan outside the identified desert dust event may differ in source characteristics and ice-nucleating efficiency from those of the highly active desert dust represented by the D15 parameterization. The apparent overestimation by D15 during pollution episodes implies that these coarse particles may possess lower ice-nucleating efficiency than the highly active desert dusts represented by the parameterization."

Lines 298-300: “This comparison suggests that, under the East Asian desert dust transport regime, typical atmospheric aging does not significantly modify the surface ice-nucleating activity of mineral dust, and that mineralogical composition and particle-size characteristics are likely more important determinants.” I found this highly speculative.

Response: The original wording was too strong because the present study did not directly characterize the aging state of the transported dust particles or the specific atmospheric aging processes involved. We have revised this part to separate the observational comparison from any inference about atmospheric aging. The revised text now states that the n_s values of the Taiyuan dust samples broadly overlap with the Beijing dust parameterization, suggesting that strong ice-nucleating activity was retained during the Taiyuan dust episode. We also cite Chen et al. (2023), who reported observational evidence that atmospheric chemical modification did not suppress the ice nucleation activity of East Asian dust, as relevant context. However, this citation is not used to claim that the present dataset directly demonstrates an aging effect. We now explicitly state that the comparison in this study should not be interpreted as direct evidence for, or a quantification of, the effect of atmospheric aging.

Changes in manuscript:

Section 3.3: “As shown in Fig. 3, the Beijing dust parameterization broadly overlaps with the n_s values of the dust samples in this study across the measured temperature range. This comparison is consistent with Chen et al. (2023), who reported observational evidence that atmospheric chemical modification did not suppress the ice nucleation activity of East Asian dust. However, because the present study did not directly characterize the aging state of the dust particles or the specific aging processes involved, this comparison is used only to suggest that strong ice-nucleating activity was retained during the Taiyuan dust episode, rather than to demonstrate or quantify the effect of atmospheric aging.”

Lines 359-360: “provide a framework for evaluating the contributions of local anthropogenic emissions versus naturally transported particles.” I don’t think this was clearly demonstrated.

Response: We have revised this sentence to a more cautious description of the observational value of the present dataset.

Changes in manuscript:

Conclusions: “The results provide observational constraints on wintertime INP abundance and variability in an industrial urban atmosphere.”

Lines 366-367: “suggesting that typical aging processes do not substantially suppress the INP efficiency of East Asian mineral dust.” No evidence was provided for this.

Response: We have removed this statement and revised the text to a more cautious description based only on the observed strong ice-nucleating activity of the transported desert dust during the dust event.

Changes in manuscript:

Conclusions: “Comparison with previously reported dust parameterizations indicates

that the transported desert dust retained strong ice-nucleating activity during the dust transport episode.”

Line 375: “The findings show that long-range desert dust transport is the decisive driver of INP enhancements in Taiyuan” This is a very strong conclusion from a single dust event.

Response: We have revised this sentence to a more cautious formulation limited to the present observations.

Changes in manuscript:

Conclusions: “Overall, the observations from this winter campaign indicate that the strongest INP enhancements in Taiyuan were associated with long-range desert dust transport, whereas the remaining INP variability during the non-desert-dust periods reflected more complex influences that were not resolved by the major PM_{2.5} source factors considered here. These results provide observational constraints for understanding wintertime urban INPs and for improving their representation in chemical transport and climate models.”

Lines 378-379: “These observations help clarify the relative contributions of natural and anthropogenic sources to INPs”. I am not fully convinced about this.

Response: We have revised this sentence to a more cautious statement emphasizing the observational constraints provided by the present study.

Changes in manuscript:

Conclusions: “These results provide observational constraints for understanding wintertime urban INPs and for improving their representation in chemical transport and climate models.”

Section 3.3. I suggest adding a Table similar to Table S1 but for the dust period

Response: We considered adding a table analogous to Table S1 for the dust period. However, the identified desert dust event in this study was limited to a short time window (5–9 December), resulting in too few samples for a robust Pearson correlation analysis comparable to Table S1. We were concerned that a dust-period correlation table based on such a limited sample size would be statistically unstable and potentially overinterpreted. We therefore chose not to add a separate correlation table for the dust period. Instead, the dust event is characterized in the manuscript using its time evolution, backward trajectories, coarse-particle enhancement, PM₁₀ increase, and the corresponding N_{INP} and ns behavior.

Figure 1. Panel a is not easy to follow. Perhaps using more contrasting colors could enhance its readability

Response: We revised the way special periods are indicated, replacing the original shaded highlighting with dashed boundary lines and colored arrows. This improves readability without obscuring the plotted data.

Changes in manuscript:

Figure 1:

The original shaded highlighting of special periods has been replaced by dashed boundary lines and colored arrows to improve readability and make the event periods easier to follow.

Figure 3. add n_s values for literature studies on urban particles and dust

Response: We have revised Fig. 3 by adding n_s values for urban anthropogenic dust particles reported by Chen et al. (2024), in addition to the existing desert-dust and K-feldspar parameterizations and the Beijing dust-event observations. This added reference helps place the Taiyuan n_s values in the context of both transported desert dust and urban dust-related particles.

Changes in manuscript:

Figure 3 and its caption were revised to include literature n_s values for urban anthropogenic dust particles reported by Chen et al. (2024).

“The light-pink dashed line denotes the median trend of n_s values for urban anthropogenic dust particles in Beijing, digitized from Chen et al. (2024).”

Figure 4. This data adds little to the discussion.

Response: We have revised the accompanying text to clarify that Fig. 4 provides the temporal context for the PMF– N_{INP} correlation analysis by showing the daily contributions of the major PM_{2.5} source factors during the non-desert-dust periods. This helps interpret the subsequent correlation results, because secondary aerosols and coal/traffic emissions dominated the PM_{2.5} mass during much of the non-desert-dust period but did not show significant covariation with N_{INP} . We therefore retained Fig. 4 in the main text but revised the discussion and caption to make its purpose clearer.

Changes in manuscript:

Section 3.4: “The reconstructed daily PM_{2.5} contributions (Fig. 4) provide the temporal context for the subsequent PMF– N_{INP} correlation analysis, showing that secondary aerosols and coal/traffic emissions dominated the PM_{2.5} mass during much of the non-desert-dust period, whereas fireworks emissions became prominent during the New Year period.”

Figure 4 caption: “Daily PM_{2.5} source contributions resolved using the PMF model, shown together with the corresponding N_{INP} at –20 °C during the non-desert-dust periods. Data from the identified desert dust event are not shown.”

Technical comments:

Line 15: Replace “campaign in Taiyuan” by “campaign in Taiyuan (China)”

Response: Revised as suggested.

Line 69: Replace “dust in cities” by “dust in urban environments”

Response: Revised as suggested.

Line 71: “urban and peri-urban regions (Chen et al., 2024)”. I suggest adding other references.

Response: After rechecking the relevant literature, we found that direct field evidence for anthropogenic dust as an atmospheric INP source in urban or peri-urban environments remains limited. Therefore, instead of adding less directly relevant

references, we revised the sentence to make this limitation explicit and retained Chen et al. (2024) as the most directly relevant reference.

Changes in manuscript:

Introduction: “Furthermore, the influence of dust in urban environments is not limited to long-range natural transport; anthropogenic dust may also contribute to atmospheric INPs in some urban and peri-urban regions, although direct field evidence remains limited (Chen et al., 2024).”

Line 35: “thermal activation behavior” What does it mean?

Response: We agree that the phrase “thermal activation behavior” was unclear, and we have revised it to a more specific wording.

Changes in manuscript:

“thermal activation behavior” has been revised to “temperature dependence of INP activation”.

References

Hu, Y., Tian, P., Huang, M., Bi, K., Schneider, J., Umo, N. S., Ullmerich, N., Höhler, K., Jing, X., Xue, H., Ding, D., Liu, Y., Leisner, T., and Möhler, O.: Characteristics of ice-nucleating particles in Beijing during spring: A comparison study of measurements between the suburban and a nearby mountain area, *Atmospheric Environment*, 293, 119451, <https://doi.org/10.1016/j.atmosenv.2022.119451>, 2023.

Chen, J., Wu, Z., Chen, J., Reicher, N., Fang, X., Rudich, Y., and Hu, M.: Size-resolved atmospheric ice-nucleating particles during East Asian dust events, *Atmospheric Chemistry and Physics*, 21, 3491–3506, <https://doi.org/10.5194/acp-21-3491-2021>, 2021.

Chen, J., Wu, Z., Meng, X., Zhang, C., Chen, J., Qiu, Y., Chen, L., Fang, X., Wang, Y., Zhang, Y., Chen, S., Gao, J., Li, W., and Hu, M.: Observational evidence for the non-suppression effect of atmospheric chemical modification on the ice nucleation activity of East Asian dust, *Sci. Total Environ.*, 861, 160708, <https://doi.org/10.1016/j.scitotenv.2022.160708>, 2023.

RC3

General Comments:

This manuscript reports on the results of a month-long field measurements of immersion-mode ice-nucleating particles (INPs) in Taiyuan, China, in winter (from 3 December 2023 to 14 January 2024). Since INP data obtained in cities of East Asia other than Beijing and Tokyo are limited, I think that the datasets obtained in Taiyuan are valuable for publication. However, the key conclusions of this work are essentially the same as those of previous studies in Beijing and Tokyo. For example, drastic increases in INPs caused by Asian dust events have been reported in Beijing (Zhang et al., 2022) and Tokyo (Isono et al., 1959; Tobo et al., 2020), and a weak correlation between INPs and anthropogenic aerosols (or PM_{2.5}) have been reported in Beijing (Chen et al., 2018; Bi et al., 2019; Zhang et al., 2022), respectively. For this reason, I don't think that this work meets the criterion of ACP's “Research articles”, which should report substantial new results and conclusions from scientific investigations of atmospheric properties, and processes. Instead, I would recommend this manuscript for

publication as the category of “Measurement reports” if the authors could improve the quality of the data analysis and figures and provide further discussion.

Response: We thank the reviewer for the careful reading of our manuscript and for recognizing the value of the Taiyuan INP dataset. We appreciate the reviewer’s concern that some of the broad features observed here, such as enhanced INPs during dust events and weak relationships with major anthropogenic aerosol components, are also consistent with previous studies in Beijing and Tokyo. At the same time, we believe that the present study provides useful new observational constraints from a heavily industrialized semi-arid city in North China, where comparable wintertime urban INP observations remain very limited. Following the reviewer’s suggestion, we have substantially improved the data analysis, figures, and discussion throughout the revised manuscript. Detailed responses are provided below.

The title of new version manuscript was changed to be “Measurement report: Dust Transport and Local Anthropogenic Emissions Differently Shape Atmospheric Ice-Nucleating Particles in an Industrial Urban Atmosphere”

Specific Comments:

1) I doubt if the ice-nucleation active site density (n_s) values are calculated appropriately, since the size range used for the calculation of the total surface area (A) is not provided in Line 158. If A was calculated based on SMPS data only (3 to 453 nm) as written in Lines 171-172, both the A and n_s values should be recalculated by combining SMPS data and Optical Particle Counter data (0.5 to 20 μm).

Response: We thank the reviewer for this important comment. In the actual n_s calculation, the total particle surface area A was derived from the combined particle size distributions measured by the SMPS and OPC, rather than from SMPS data alone. Thus, the contribution of particles larger than 0.5 μm was already included in the calculation, and recalculation of n_s was not required. We have revised the manuscript to clarify this point explicitly and to better describe how the merged SMPS–OPC size distribution was used to estimate A for n_s calculation.

Changes in manuscript:

Section 2.2.1: “The OPC data were combined with the SMPS measurements to construct a merged particle size distribution (SMPS: 3–453 nm; OPC: 0.5–20 μm), which was further used to estimate the total particle surface area for n_s calculation.”

Section 2.2.3: “For $n_s(T)$ calculation, the particle surface area was estimated from the merged SMPS–OPC size distribution expressed in mobility diameter.”

Section 3.3: “The n_s values discussed here were calculated from the merged SMPS–OPC size distribution, thereby accounting for contributions from coarse particles in the surface-area estimate.”

2) Dust INP parameterization of DeMott et al. (2015) was designed to calculate INP number concentrations related to the number concentrations of dust particles larger than 0.5 μm and not those of total aerosol particles larger than 0.5 μm . If the INP number concentrations for D15 (–20°C) shown in Figure 1a were calculated using the number concentrations of total aerosol particles larger than 0.5 μm , it would not be an

appropriate use.

Response: We agree that the DeMott et al. (2015) parameterization was originally developed for desert dust aerosol and, strictly speaking, is most appropriate when applied to dust particles larger than 0.5 μm rather than to total aerosol particles larger than 0.5 μm . In this study, the D15 values shown in Fig. 1a were calculated using the observed particle number concentrations larger than 0.5 μm , because dust-only number concentrations were not available. We therefore do not treat D15 here as a strict quantitative predictor, especially outside the identified dust episode. Instead, it is retained mainly as a dust-based reference to compare the temporal variation of the observations with dust-dominated behavior. We have clarified this point explicitly in the revised manuscript.

Changes in manuscript

Section 3.3: “Figure 1a also compares the measurements with the DeMott et al. (2015) parameterization (D15), which links INP concentrations to the number concentration of particles larger than 0.5 μm and was originally developed for desert dust aerosol. In this study, the D15 values were calculated using the observed particle number concentrations larger than 0.5 μm and are therefore shown mainly as a dust-based reference. Accordingly, the comparison is most meaningful for the identified dust episode, whereas for the remaining periods it should be interpreted with caution.”

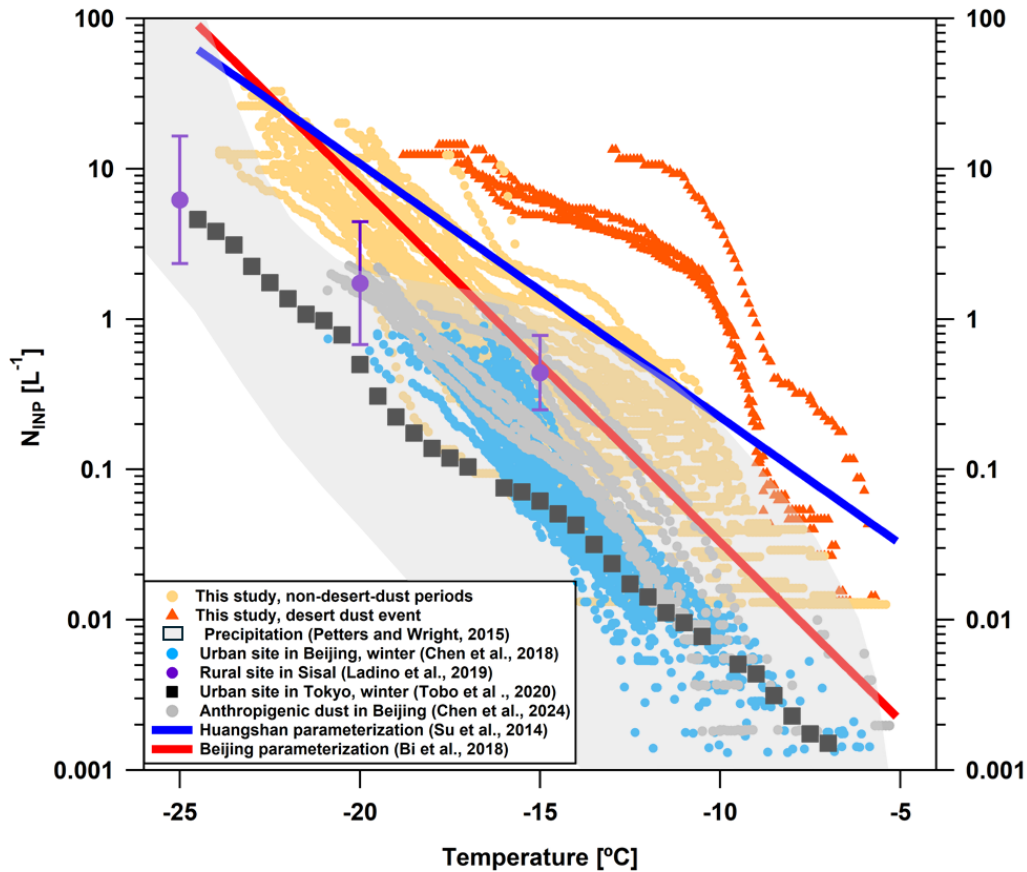
Figure 1 caption: “In panel (a), gray triangles (D15(-20°C)) represent values calculated using the INP parameterization scheme proposed by DeMott et al. (2015). Here, they are shown mainly as a dust-based reference to compare the day-to-day temporal variation between parameterized and observed INP concentrations.”

3) Figure 2: For reference data, the period of the data sampling should be indicated in addition to the location. In this regard, although the period of the INP data in Tokyo (Tobo et al., 2020) shown in Figure 2 is unclear (since the period is not indicated in this figure), it would be appropriate to show the data collected in winter for comparison. In addition, I strongly suggest including the INP data in Beijing reported by Chen et al. (2018), because they reported the INP data in Beijing during winter, which were measured using the same technique as this study.

Response: We thank the reviewer for this helpful suggestion. We have revised Fig. 2 by adding wintertime urban INP observations in Beijing measured by INDA during November–December 2016 (Chen et al., 2018). We also revised the Tokyo comparison to use wintertime data from December 2016 to February 2017 reported by Tobo et al. (2020). The figure caption has been updated accordingly.

Changes in manuscript:

Figure 2 and its caption were revised to include wintertime Beijing data from Chen et al. (2018) and wintertime Tokyo data from Tobo et al. (2020), with the sampling periods indicated.



4) It seems that the INP number concentrations in Taiyuan are somewhat higher than those in Beijing (Chen et al., 2018) and Tokyo (Tobo et al., 2020) during winter, even though these data have been obtained using essentially the same technique (i.e., cold-stage-based technique). Please discuss possible reasons for higher INP number concentrations in Taiyuan and possible differences (e.g., location and possible major sources of INPs) between Taiyuan and these urban sites in East Asia. In this regard, I would like to suggest comparing available ambient aerosol data (e.g., number concentrations of aerosol particles larger than 0.5 μm , $\text{PM}_{2.5}$, PM_{10} , chemical composition) between Taiyuan and other sites.

Response: We thank the reviewer for this helpful suggestion. In the revised manuscript, we have updated Fig. 2 to include the wintertime Beijing data of Chen et al. (2018) and wintertime Tokyo data from Tobo et al. (2020), which provide more directly comparable cold-stage-based urban observations. We have also added a discussion of the possible reasons for the higher N_{INP} levels in Taiyuan. The most pronounced enhancement was associated with the identified 5–9 December desert dust intrusion, as supported by back trajectories, elevated PM_{10} , and enhanced coarse-mode particle concentrations. During the non-desert-dust periods, the relatively high N_{INP} levels may still reflect the semi-arid setting of Taiyuan, its proximity to the Loess Plateau and northwestern dust source regions, and the influence of local dust-associated and resuspended coarse particles. In addition, differences in sampling size range should be considered, because the Taiyuan INP filters were collected without a size-selective inlet and can be regarded as approximately TSP, whereas the Beijing winter data of Chen et

al. (2018) were based on PM_{2.5} filter samples. We have framed these explanations as plausible factors rather than a quantitative source attribution, because directly comparable aerosol datasets are not available for all sites.

Changes in manuscript:

Section 3.2: We added a discussion of possible reasons for the higher N_{INP} levels in Taiyuan compared with wintertime observations in Beijing and Tokyo, including the semi-arid setting of Taiyuan, its proximity to the Loess Plateau and northwestern dust source regions, the persistence of coarse-mode particles, and differences in sampling size range among studies.

Technical Comments:

5) Lines 39-41: Given that some recent studies render deposition nucleation unlikely for the formation of ice clouds in the atmosphere and “pore condensation and freezing” is a more likely mechanisms for atmospheric ice nucleation below water saturation (e.g., Marcolli, 2014; David et al., 2019), it might be appropriate to describe “pore condensation and freezing” in addition to the four mechanisms.

Response: We agree that pore condensation and freezing (PCF) should be acknowledged in this part of the Introduction. We have therefore added a brief sentence on PCF and cited the suggested references.

Changes in manuscript:

Introduction: “In addition, recent studies have suggested that under water-subsaturated conditions, pore condensation and freezing (PCF) may provide a more realistic description of atmospheric ice nucleation than classical deposition nucleation in some cases (Marcolli, 2014; David et al., 2019).”

6) Lines 51-52: Please indicate the references regarding this explanation. In addition, the authors would need to explain that some studies have indicated the negligible contribution of soot particles to INPs in the immersion mode (e.g., Kanji et al., 2020).

Response: We agree that the original explanation required clearer supporting references and that the discussion of soot should be more nuanced. We have therefore revised this part of the Introduction by adding references for anthropogenic ice-active particles and by clarifying that soot particles may contribute negligibly to atmospheric INPs in the immersion mode, following Kanji et al. (2020).

Changes in manuscript:

Introduction: “However, this observation contrasts with laboratory and field evidence showing that some anthropogenic particles can exhibit immersion-mode ice-nucleating activity. These include certain fly ash, metallic/mineral particles, and other industrial dusts associated with coal combustion and manufacturing activities (Toll et al., 2024). By contrast, the contribution of soot particles to atmospheric INPs in the immersion mode has been suggested to be negligible in many cases (Kanji et al., 2020).”

7) Line 83: As far as I check the data presented in this manuscript, it seems that the INP data used here are limited to the temperature regime from about -25°C to -5°C.

Response: In this study, the freezing experiments were continued until all droplets were

frozen. However, because most samples were already nearly or completely frozen by about $-25\text{ }^{\circ}\text{C}$, the INP data presented in this manuscript mainly span approximately $-25\text{ }^{\circ}\text{C}$ to $-5\text{ }^{\circ}\text{C}$. We have revised the text accordingly.

Changes in manuscript:

Introduction: “In this study, the number concentration of immersion mode INP (N_{INP}) and the number of active sites per unit surface area of INPs (n_s) were measured under mixed-phase cloud conditions. The freezing experiments were continued until all droplets were frozen, but because most samples were already fully or nearly fully frozen by about $-25\text{ }^{\circ}\text{C}$, the INP data presented and discussed in this study mainly span the temperature range from about $-25\text{ }^{\circ}\text{C}$ to $-5\text{ }^{\circ}\text{C}$.”

8) Lines 160-169: If the confidential intervals for the INP number concentrations were calculated the error bars should be indicated in the INP data in the figures.

Response: We thank the reviewer for this comment. Because the main figures contain dense temperature-resolved data, adding point-by-point confidence intervals would reduce readability. We have therefore added a new supplementary table reporting N_{INP} values and corresponding 95% confidence intervals at selected temperatures. We also added a sentence in Section 3.1 referring readers to this table.

Changes in manuscript:

Section 3.1: “The corresponding 95% confidence intervals for N_{INP} at selected temperatures are provided in Table S3.”

Supplement, Table S3: “**Table S3.** N_{INP} values and corresponding 95% confidence intervals at selected temperatures. Values are shown as N_{INP} (lower–upper), in units of L^{-1} . n.q. = not quantified because complete freezing occurred before the corresponding temperature.”

9) Line 227: What do you mean by “secondary pollution”?

Response: Here, “secondary pollution” was intended to refer to pollution episodes characterized by enhanced secondary inorganic and organic aerosol accumulation. We have revised it accordingly.

Changes in manuscript:

Section 3.1: “secondary pollution periods” has been revised to “pollution episodes characterized by enhanced secondary inorganic and organic aerosol accumulation”.

10) All Figures: The size of the figure legends are too small and it is hard to read. Please increase the font size.

Response: We have checked all figures and adjusted the legend font sizes where feasible. Because several figures contain dense datasets and multiple reference curves, large increases in legend size would obscure the plotted data. We therefore made moderate adjustments to balance legend readability and data visibility, and exported the revised figures at high resolution.

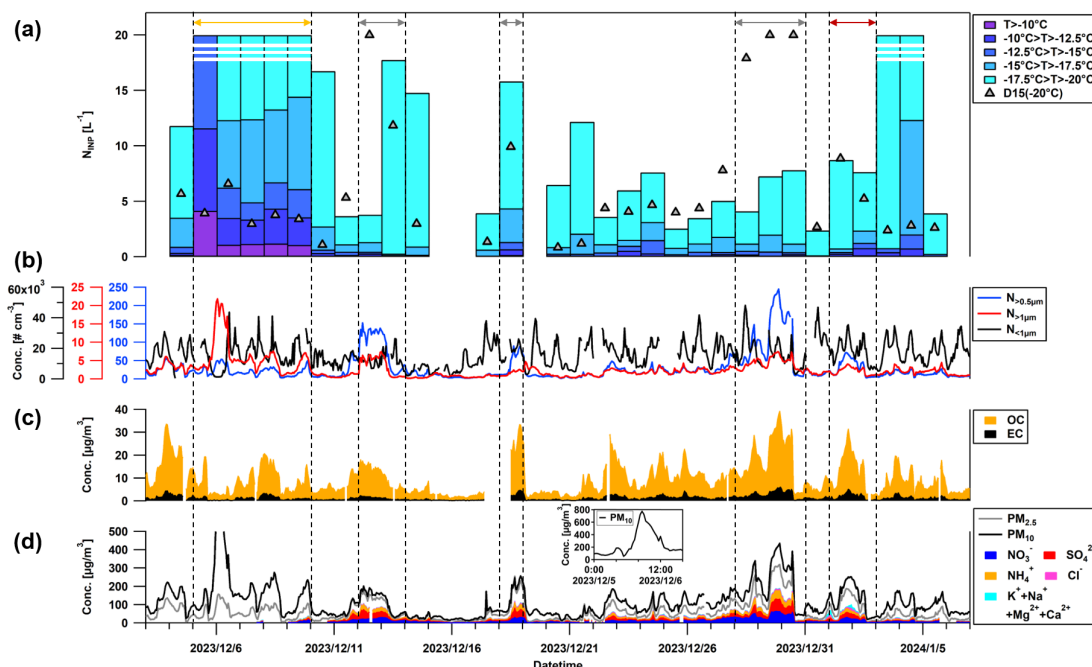
11) All Figures: Since similar colors are used in the figures (especially, Figures 1a, 1b, 1d, and 2), it is hard to see the difference in color. Please use colors and symbols that

are easier to distinguish.

Response: We have carefully re-examined the figures, especially Figs. 1a, 1b, 1d, and 2, and revised the color and symbol schemes as much as possible. Because these figures contain multiple time series, particle-size categories, and reference datasets, completely avoiding similar colors while keeping all information visible is challenging. Nevertheless, we have made targeted revisions to improve visual clarity. In Fig. 1, the semi-transparent colored regions were replaced by clearer lines and arrows to reduce overlap with the data. In Fig. 2, the colors and symbols for different datasets were adjusted to better distinguish the present observations, wintertime urban observations, anthropogenic dust samples, and parameterization curves.

Changes in manuscript:

Figures 1 and 2 were revised to improve the distinguishability of colors and symbols. In Fig. 1, colored shaded regions were replaced by lines and arrows, and in Fig. 2, the colors and symbols of the different datasets were adjusted to improve visual separation.



12) Figure 1a: What do the three white horizontal lines indicate?

Response: The three white horizontal lines in Fig. 1a indicate samples for which all droplets had already frozen before reaching the colder temperature intervals represented by the remaining colors. Under such conditions, the cumulative N_{INP} can no longer be quantified as a finite value by the droplet-freezing method, because f_{ice} has reached 1. We have clarified this explicitly in the revised figure caption.

Changes in manuscript:

Figure 1 caption: “In panel (a), white horizontal lines indicate samples for which all droplets had already frozen before the colder temperature intervals were reached. Under these conditions, the cumulative N_{INP} exceeded the quantifiable range of the assay and therefore could not be plotted as a finite value.”

13) Figure 1a legend: The symbol “<” should be “>”.

Response: The symbol in the legend has been corrected.

Changes in manuscript:

Figure 1a legend: The symbol “<” has been corrected to “>”.

14) Figure 1b: What is the lower limit for size range of $N_{<1\mu\text{m}}$?

Response: We have therefore revised the caption to specify the diameter ranges used for $N_{<1\mu\text{m}}$, $N_{>0.5\mu\text{m}}$, and $N_{>1\mu\text{m}}$.

Changes in manuscript:

Figure 1 caption:

“In panel (b), $N_{<1\mu\text{m}}$ refers to particles from 3 nm to 1 μm , whereas $N_{>0.5\mu\text{m}}$ and $N_{>1\mu\text{m}}$ denote the concentrations integrated over 0.5–20 μm and 1–20 μm , respectively.”

15) Figure 1d: What does a small figure in Figure 1d indicate? (it is too small, anyway.)

Response: The inset in Fig. 1d is intended to highlight the extreme PM_{10} peak around 5–6 December that is not fully visible in the main panel. We have clarified this directly in the figure and improved its readability.

Changes in manuscript:

Figure 1 caption:

“The inset in panel (d) shows the full PM_{10} peak on 5–6 December.”

16) Figure 1d: I can’t see the ion data before ~7 December 2023. Is it due to their extremely low values?

Response: The ion data before approximately 7 December 2023 are not visible in Fig. 1d because the corresponding MARGA measurements were unavailable during the beginning of the campaign. We agree that this was not clearly stated in the original manuscript and have clarified it in the revised figure caption.

Changes in manuscript:

Figure 1 caption:

“Ion data before approximately 7 December 2023 were unavailable and are therefore not shown in panel (d).”

17) Figure 2: The data for “Urban site in New Delhi (Wagh et al., 2021)” would not be appropriate for this figure, because they reported INP data in the deposition mode and not in the immersion mode.

Response: We agree that the New Delhi dataset reported by Wagh et al. (2021) is not directly comparable to the present study, because it was obtained in the deposition mode rather than the immersion mode. We have therefore removed this dataset from Fig. 2 and revised the associated text accordingly.

Changes in manuscript:

Figure 2 and its caption were revised by removing the New Delhi dataset. The direct comparison with New Delhi in Section 3.2 was also removed.

References:

Bi et al. (2019), <https://doi.org/10.1029/2019JD030609>

Chen et al. (2018), <https://doi.org/10.5194/acp-18-3523-2018>
 David et al. (2019), <https://doi.org/10.1073/pnas.1813647116>
 DeMott et al. (2015), <https://doi.org/10.5194/acp-15-393-2015>
 Isono et al. (1959), https://doi.org/10.2151/jmsj1923.37.6_211
 Kanji et al., (2020), <https://doi.org/10.1029/2019GL086764>
 Marcolli (2014), <https://doi.org/10.5194/acp-14-2071-2014>
 Tobo et al. (2020), <https://doi.org/10.1029/2020JD033658>
 Wagh et al. (2021), <https://doi.org/10.1016/j.atmosres.2021.105693>
 Zhang et al. (2022), <https://doi.org/10.5194/acp-22-7539-2022>

RC4

General comments:

Atmospheric ice-nucleating particles (INPs) play a critical role in cloud formation, cloud radiative properties and precipitation through heterogeneous ice nucleation, yet the importance of INPs from anthropogenic sources remains poorly understood. This manuscript investigated INP concentrations (N_{INP}) and ice nucleation active site density (ns) of immersion mode INPs in Taiyuan, China based on one month-long winter field observations. The results indicated that the temporal variability of INP concentrations is dominated by episodic natural desert dust transport, and the direct impact of anthropogenic emissions on INP loading remains limited. Namely, this study implied that in heavily polluted environments, natural dust rather human activity governs cloud freezing processes. These findings would offer observational constraints for improving INP parameterizations in chemical transport and climate models.

In general, I think the research goal of this manuscript is clear, and the structure and results are reasonable. Therefore, I recommend this manuscript is accepted and published in the journal of *EGU*sphere after some revisions. The authors are advised to consider the following comments.

Response: We are very grateful to the reviewers for their constructive comments. In response to these comments, we have provided point-by-point replies and made corresponding revisions throughout the manuscript.

Specific comments:

(1) Page 1, Abstract, Lines 18-19: ‘CI: 6.64-8.41, CI: 1.58-1.99’.

Comment: When an English abbreviation first appears in the manuscript, please provide its full form.

Response: In the revised manuscript, we have replaced “CI” with “confidence interval (CI)” in the Abstract.

Changes in manuscript:

Page 1, Abstract: “CI” has been revised to “confidence interval (CI)” at its first appearance.

(2) Page 1, Abstract, Lines 22-23; Page 6, 2.2.5 Positive matrix factorization (PMF), Lines 179-186: ‘Positive matrix factorization (PMF) identified five PM₅ sources: coal combustion and traffic emissions, industry, (anthropogenic) dust, secondary aerosols

and fireworks.’

Comment: How did the PMF method distinguish between natural dust and anthropogenic dust? Please provide a detailed description in the context. This is of great significance for the analysis of how the two different types of dust affect the INPs in the manuscript.

Response: In this study, PMF resolved a dust-related factor characterized by crustal elements such as Ca and Fe, but PMF alone was not used to directly distinguish transported desert dust from local dust-associated particles. The distinction was made using additional evidence, including backward trajectories, coarse-mode particle enhancement, PM₁₀ variation, and event classification of the 5–9 December desert dust event. We have therefore revised the wording throughout the manuscript from “anthropogenic dust” to “dust-related factor” or “local dust-associated particles” where appropriate. We now clarify that PMF was used to identify a dust-related PM_{2.5} source factor and to assess its covariation with N_{INP} during non-desert-dust periods, rather than to directly separate natural and anthropogenic dust sources.

Changes in manuscript:

Abstract: “Positive matrix factorization (PMF) identified five PM_{2.5} sources: industrial emissions, dust-related particles, secondary aerosols, coal combustion and traffic emissions and fireworks.”

Section 3.4: “Data from the identified desert dust event (5–9 December) are not shown in Fig. 4, because the PMF–N_{INP} correlation analysis focused on non-desert-dust periods. The dust-related factor exhibits characteristic signatures of dust-associated particles, including high loadings of Ca and Fe. During the non-desert-dust periods, this factor is interpreted as mainly reflecting local dust-associated particles, such as road dust, fugitive dust from industrial and construction activities, and resuspended soil. This dust-related PM_{2.5} factor showed no significant correlation with N_{INP} during the non-desert-dust periods (Table 1), indicating that local dust-associated PM_{2.5} variability did not explain N_{INP} variability outside the identified long-range desert dust event.”

(3) Page 1, Abstract, Lines 26-27 and in the context: ‘Observed N_{INP} variability is likely governed by the interplay of episodic coarse-mode inputs and atmospheric processing rather than a single dominant source’.

Comment: In the manuscript, the authors only provided one month-long winter field observations in Taiyuan, which is insufficient to confirm this conclusion. That is, this conclusion is difficult to convince the readers.

Response: Our intention was to summarize the observed pattern during this campaign, rather than to draw a general mechanistic conclusion beyond the temporal and spatial scope of the present dataset. In the revised manuscript, we have therefore softened this statement in the Abstract and in the main text. Specifically, we now describe the observations more cautiously by emphasizing that, during this winter campaign in Taiyuan, N_{INP} variability was characterized by strong episodic enhancement during the desert dust event, whereas the variability during non-desert-dust periods could not be explained by any single major PM_{2.5} source factor. We have also revised related overgeneralized statements in the Discussion and Conclusions accordingly.

(4) Page 3, Lines 69-71: ‘Furthermore, the influence of dust in cities is not limited to long-range natural transport; anthropogenic dust-such as agricultural operations, construction, and other urban processes-can also contribute substantially to atmospheric INPs in urban and peri-urban regions (Chen et al., 2024)’.

Comment: The main conclusion of this manuscript seems to be in contradiction to the results obtained by Chen et al. (2024). In order to facilitate readers' understanding, please provide a reasonable explanation in the context.

Response: Our intention in citing Chen et al. (2024) in the Introduction was to note that anthropogenic dust has been reported as a potentially important source of urban INPs during non-dust periods, rather than to suggest that the same source necessarily dominated the present observations in Taiyuan. We agree, however, that the original wording in the Discussion, especially the sentence “This implies that the INPs observed in Taiyuan may also be influenced by anthropogenic dust particles,” was too speculative and may have created an apparent contradiction with our main conclusion.

Changes in manuscript:

(1) Introduction: “Furthermore, the influence of dust in urban environments is not limited to long-range natural transport; anthropogenic dust may also contribute to atmospheric INPs in some urban and peri-urban regions, although direct field evidence remains limited (Chen et al., 2024).”

(2) Section 3.2: “This comparison suggests that the relative importance of anthropogenic dust and transported desert dust may vary with urban environments and observation period. In this winter campaign in Taiyuan, the strongest INP enhancements were associated with the identified desert dust event.”

(5) Page 4, Lines 100-102: ‘In addition, a two-channel sampler was used to collect particles on filters for INP measurement. INP filter samplesusing a two-channel sampler without a cyclone, from 4 December 2023 to 5 January 2024’.

Comment: What is the meaning of ‘using a two-channel sampler without a cyclone’? Please provide the measurement particle size ranges of INP in the context, which is vital to the analysis of the results in this manuscript.

Response: Here, “without a cyclone” means that no size-selective inlet (cyclone and other impactors) was installed upstream of the filters. Therefore, no defined aerodynamic cut-off was applied during INP sampling, and the collected aerosol can be regarded as approximately total suspended particles. We have revised the manuscript accordingly.

Changes in manuscript:

Section 2.1: “In addition, INP filter samples were collected using a two-channel sampler equipped with two parallel filter channels from 4 December 2023 to 5 January 2024. Polycarbonate filters (47 mm, Nuclepore Track-Etch Membrane, 0.2 μm pore size, Whatman) were used for sampling. The sampler was operated without a size-selective inlet.”

(6) Page 9, Lines 251-253: ‘Chen et al. (2024) found that anthropogenic dust particles,

such as road dust influenced by traffic emissions, are important INP sources in the urban atmosphere. This implies that the INPs observed in Taiyuan may also be influenced by anthropogenic dust particles’.

Comment: The conclusion of ‘This implies that the INPs observed in Taiyuan may also be influenced by anthropogenic dust particles’ seems to be in contradiction to the main conclusion of the manuscript ‘In summary, long-range mineral dust transport is the decisive driver of INP enhancements, while traditional anthropogenic fine aerosols contribute minimally’ (**Abstract, Lines 25-26**). Please provide a reasonable explanation in the context.

Response: We thank the reviewer for this important comment. We agree that the original sentence, “This implies that the INPs observed in Taiyuan may also be influenced by anthropogenic dust particles,” was too speculative and could be read as inconsistent with the main conclusion of the manuscript. Our intention was not to infer anthropogenic dust influence in Taiyuan directly from Chen et al. (2024), but rather to note that anthropogenic dust has been reported as an important urban INP source in some environments.

To avoid this ambiguity, we have removed the above sentence and revised the surrounding discussion. The revised text now states more cautiously that Chen et al. (2024) and the present study together suggest that the relative importance of anthropogenic dust and transported desert dust may vary among urban environments and observation periods. In the present winter campaign in Taiyuan, the strongest INP enhancements were associated with the identified desert dust event, whereas the remaining variability during non-desert-dust periods could not be uniquely attributed to anthropogenic dust based on the present dataset.

Changes in manuscript:

Same as (4) Page 3, Lines 69-71: (2)

(7) Page 11, Lines 298-300: ‘This comparison suggests that, under East Asian desert dust transport regime, typical atmospheric aging does not significantly modify the surface ice-nucleating activity of mineral dust, and that mineralogical composition and particle-size characteristics are likely more important determinants’.

Comment: The author didn’t provide direct and sufficient evidences in the manuscript to prove this conclusion. Please revise this conclusion or provide sufficient evidence in the manuscript. For instance, ‘typical atmospheric aging does not significantly modify the surface ice-nucleating activity’, the authors didn’t provide the ‘typical atmospheric aging processes’ or ‘aging mineral dust aerosol’ in the context.

Response: The original wording was too strong because the present study did not directly characterize the aging state of the transported dust particles or the specific atmospheric aging processes involved. We have revised this part to separate the observational comparison from any inference about atmospheric aging. The revised text now states that the ns values of the Taiyuan dust samples broadly overlap with the Beijing dust parameterization, suggesting that strong ice-nucleating activity was retained during the Taiyuan dust episode. We also cite Chen et al. (2023), who reported observational evidence that atmospheric chemical modification did not suppress the ice

nucleation activity of East Asian dust, as relevant context. However, this citation is not used to claim that the present dataset directly demonstrates an aging effect. We now explicitly state that the comparison in this study should not be interpreted as direct evidence for, or a quantification of, the effect of atmospheric aging.

Changes in manuscript:

(1) Section 3.3: “As shown in Fig. 3, the Beijing dust parameterization broadly overlaps with the n_s values of the dust samples in this study across the measured temperature range. This comparison is consistent with Chen et al. (2023), who reported observational evidence that atmospheric chemical modification did not suppress the ice nucleation activity of East Asian dust. However, because the present study did not directly characterize the aging state of the dust particles or the specific aging processes involved, this comparison is used only to suggest that strong ice-nucleating activity was retained during the Taiyuan dust episode, rather than to demonstrate or quantify the effect of atmospheric aging.”

(3) Conclusions: “Comparison with previously reported dust parameterizations indicates that the transported desert dust retained strong ice-nucleating activity during the dust transport episode.”

References

Chen, J., Wu, Z., Meng, X., Zhang, C., Chen, J., Qiu, Y., Chen, L., Fang, X., Wang, Y., Zhang, Y., Chen, S., Gao, J., Li, W., and Hu, M.: Observational evidence for the non-suppression effect of atmospheric chemical modification on the ice nucleation activity of East Asian dust, *Sci. Total Environ.*, 861, 160708, <https://doi.org/10.1016/j.scitotenv.2022.160708>, 2023.