Author Response to Referee #3
EGUSPHERE-2026-308

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 ns 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."

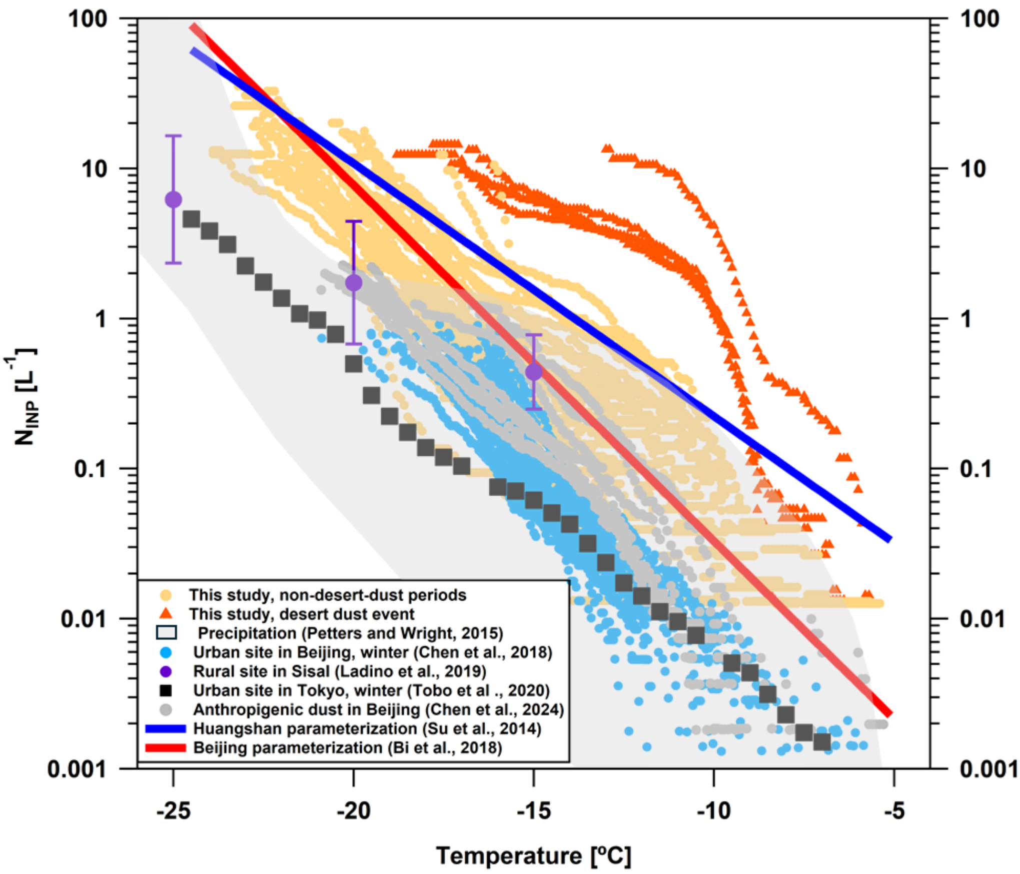
3) Figure 2: For reference data, the period of the data sampling should be indicated in addition to the location. In this regard, although the period of the INP data in Tokyo (Tobo et al., 2020) shown in Figure 2 is unclear (since the period is not indicated in this

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, $PM_{2.5}$, 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 °C, the INP data presented in this manuscript mainly span approximately −25 °C to −5 °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 °C, the INP data presented and discussed in this study mainly span the temperature range from about −25 °C to −5 °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.

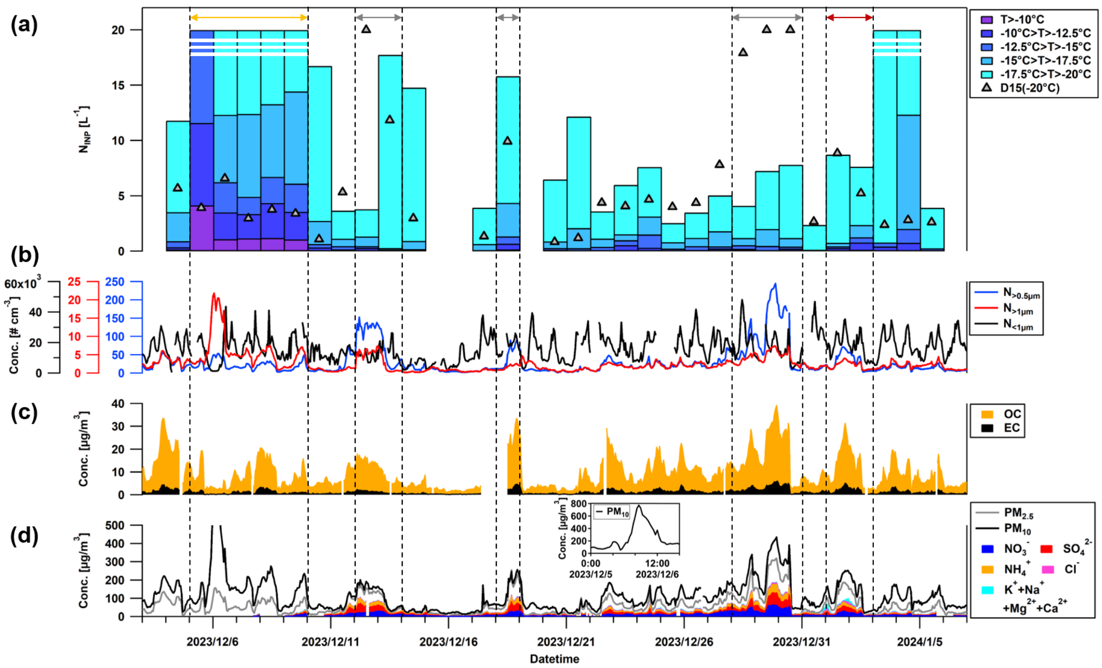
Response: We have checked all figures and adjusted the legend font sizes where feasible. Because several figures contain dense datasets and multiple reference curves, large increases in legend size would obscure the plotted data. We therefore made moderate adjustments to balance legend readability and 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 $N_{<1\mu m}$?

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

Changes in manuscript:

Figure 1 caption:

"In panel (b), $N_{<1\mu m}$ refers to particles from 3 nm to 1 μm, whereas $N_{>0.5\mu m}$ and $N_{>1\mu m}$ 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