Response to Editor and Reviewers' Comments

Thank you very much for the reviewers' comments regarding our manuscript entitled "Measurement Report: New insights into the boundary layer revolution impact on new particle formation characteristics in three megacities of China" (Manuscript ID: EGUSPHERE-2025-3637). We appreciate the valuable feedback provided by the reviewers, which has significantly contributed to enhancing the quality of our manuscript. In response, we have meticulously addressed each comment in a systematic manner, ensuring that all concerns are thoroughly considered. The manuscript has been revised accordingly, with all modifications clearly highlighted in blue within the marked version for easy reference. Additionally, comprehensive responses to the editor and reviewers' comments are detailed below, demonstrating our commitment to transparency and scholarly rigor.

Reviewer Comment:

In Line 96, the authors claimed that they utilized long-term observational data sets, which is totally overstated. The data set in Beijing merely contains 408 days, which cannot be called "long-term", and let alone the data sets in other cities.

Response:

We sincerely thank the reviewer for their critical feedback, which helps us improve the clarity and rigor of our manuscript.

We acknowledge that the "long-term" may have been an overstatement in this context. Although our dataset in Beijing includes 408 effective observation days spanning over two years (from July 2017 to October 2019), we understand that in the context of atmospheric science, a "long-term" dataset often implies multi-year continuous measurements over a span of 3 years or more.

We conducted three years of observations across three cities: Beijing (2017/07/19–2019/10/09), Guangzhou (2019/10/31–2020/03/30), and Shanghai (2020/04/05–2020/06/05). Due to instrument issues, some days in Beijing lacked valid measurements, resulting in 408 days of effective data. Moreover, in previous studies, such as that by (Sun et al., 2015), one year of data was also referred to as "long-term" in their analysis of aerosol particle composition in Beijing.

Of course, the reviewer's comment is valid. To be more precise in describing the temporal coverage—especially in comparison with the shorter campaigns in Guangzhou (127 days) and Shanghai (53 days)—we will revise the term "long-term observational data" to "an extended observational dataset covering over 400 days." In the introduction and methods sections, we will clarify the actual date range for three cities to avoid potential misinterpretation.

Reviewer Comment:

In addition, the data included in the analyses are too simple (just PBLH and PNSD), leaving no room for real in-depth analysis. As far as I know, there should be data (at least trace gases) from Chinese government that are readily accessible.

Response:

We agree with the reviewer that our current analysis is mainly based on aerosol particle number size distribution (PNSD) and planetary boundary layer height (PBLH). The reason for this focus is twofold:

1. These two variables are directly and continuously observed at high temporal resolution using our own SMPS and MPL instruments, allowing us to conduct consistent physical classification of NPF events across sites.

2. Our main objective in this study was to isolate the physical influence of boundary layer development on NPF occurrence and dynamics, rather than perform a full chemical mechanism

analysis.

We appreciate the reviewer's point that broader analyses involving trace gases (e.g., SO_2, NO_x, O_3, CO) would enhance the scientific depth. In the revised manuscript, we will clarify that our data sources go beyond PBLH and PNSD, and include additional meteorological and trace gas parameters from authoritative national datasets. Specifically, we have downloaded hourly air pollutant data from the China National Environmental Monitoring Center (CNEMC), including SO_2, O_3, CO, and NO_2 concentrations, for the cities of Beijing, Guangzhou, and Shanghai, matched to the corresponding observation periods, and obtained surface meteorological parameters from the National Centers for Environmental Information (NCEI), including: air temperature, atmospheric pressure, dew point temperature, wind direction and speed, cloud cover, precipitation. For radiation, which plays a crucial role in driving photochemistry and boundary layer evolution, we incorporated long-term radiation datasets as published by (Liu et al., 2022).

We appreciate the reviewer's suggestions, and will further elaborate on the role of the PBLH in NPF using these data in the revised manuscript.

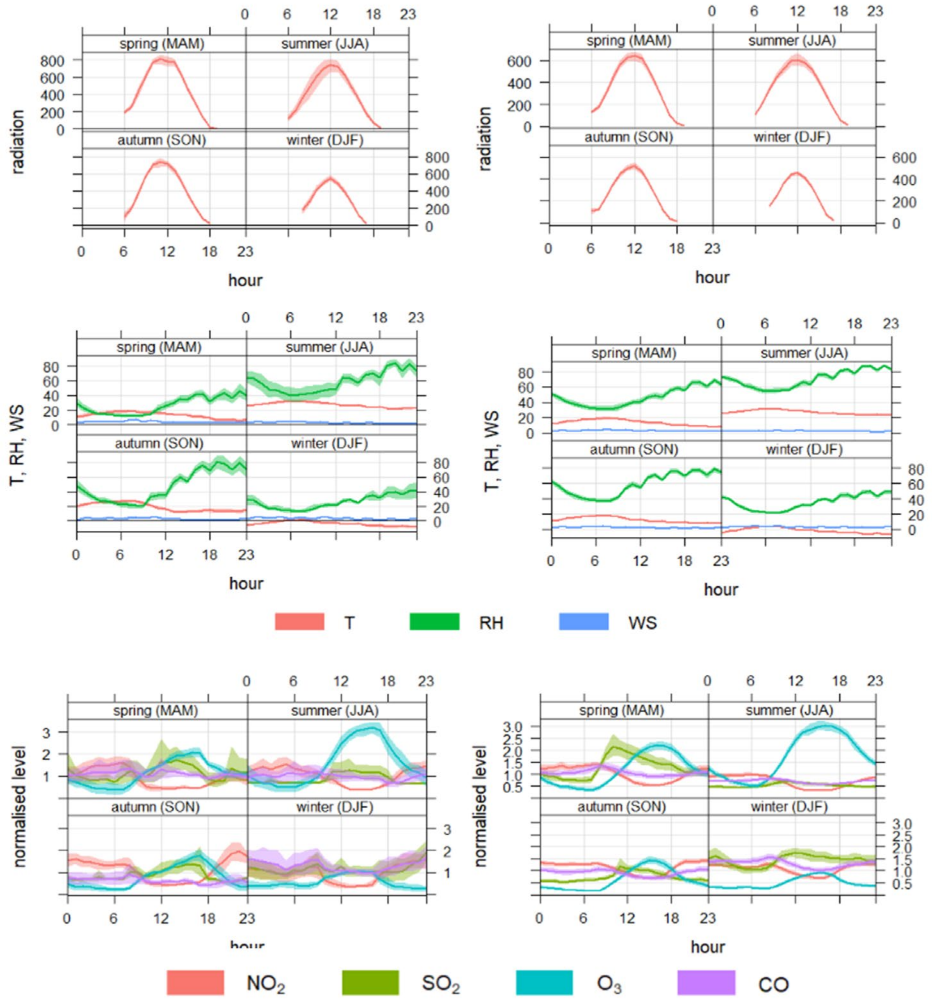

Figure R1. Seasonal variation of radiation, T, RH, WS, NO_2, SO_2, O_3 and CO.

Reviewer Comment:

Unclear statistics methods.

The PNSD shown in Fig.1 looks abnormal, please double check and specify if the contour plot is made from mean or median average data.

Response:

Thank you for your thorough and constructive comments regarding statistical methods and data presentation.

In Figure 1, the particle number concentrations in different size ranges were averaged for each season in Beijing, while seasonal averages were calculated for the entire observation periods in Guangzhou and Shanghai. There was a scaling error in the previous colorbar; we will correct it by applying a logarithmic concentration scale, which allows for clearer visualization of the variations in particle number concentrations.

The figure and caption have been updated accordingly, and a clarification has been added to the Methods section (Section 2.2) to specify the averaging method used for plotting.

Reviewer Comment:

Line 258-259. “with frequencies ranging from 3.4% to 20.0%”. It is not clear what such frequencies are calculated upon? (monthly?)

Response:

The frequencies reported as “3.4% to 20.0%” refer to monthly NPF event occurrence frequencies for Guangzhou (GZ) during the observation period.

$$NPF\ Frequency = \frac{Number\ of\ NPF\ Days\ in\ a\ Month}{Total\ Valid\ Observation\ Days\ in\ that\ Month} \times 100\%$$

We intended to convey that the overall observed NPF frequency was 10%, with monthly average frequencies ranging from 3.4% to 20.0%. To address any ambiguity caused by the original wording, we will carefully revise this part of the manuscript.

We will revise the text in Line 258–259 to clearly state:

“...with monthly NPF occurrence frequencies ranging from 3.4% to 20.0%, depending on observation coverage.”

Reviewer Comment:

In Fig.2, how was PBLH calculated for each city in different months. Are they day-time average or full-day average? Are they median or mean average? What are the variation range in PBLH?

Response:

In Figure 2, the monthly PBLH values were computed as daytime averages between 08:00 and 18:00 local time (LT), corresponding to the typical window of NPF activity. The values shown are mean values. We will specifically discuss the patterns of PBLH variation in the section related to boundary layer dynamics (Figure 3). Figure 2 only provides a preliminary overview to illustrate the monthly variation trend, while the detailed analysis will be presented in the subsequent sections.

Reviewer Comment:

Relevant to the previous comment, they authors stated that the PBLH between NPF and non-NPF days in Beijing shows significant difference. However, in Fig.3, the pattern and daily maxima of PBLH in Beijing do not very much. Is this 100 meter difference considered significant?

Response:

We also appreciate the reviewer's critical reading. In Figure 3, while the absolute difference in daily maximum PBLH between NPF and Non_NPF days in Beijing appears to be approximately 100–200 m. We agree with the reviewer that a difference of 100-200 m should not be considered a significant distinction. This discrepancy arose because we initially processed a large volume of data all at once, without accounting for seasonal variations and other influencing factors. The maximum PBLH varies from month to month and across seasons, and it also fluctuates on an hourly basis. Averaging PBLH over all days throughout the entire observation period can obscure the differences between NPF and Non-NPF events. Therefore, in the revised manuscript, we analyzed seasonal variations in Beijing and examined the hourly evolution of PBLH in different seasons.

Figure R2 presents the hourly evolution of the PBLH during NPF and Non_NPF days across different seasons in Beijing. Overall, PBLH during NPF events is generally higher than on non-event days, particularly during the key window between 09:00 and 14:00. The differences are most pronounced in spring and autumn. For example, in spring, the median PBLH at 14:00 LT on NPF days reaches approximately 1350 m, while on Non_NPF days it is only around 900 m. This corresponds well with the seasonal distribution of NPF frequency, which is highest in spring and autumn—seasons that also show the largest PBLH differences between the two types of events.

In addition, we included a third category, “shrinkage,” as a control group for comparison between NPF and Non_NPF events. Although a detailed analysis of shrinkage cases is beyond the scope of this paper, we performed statistical analysis of PBLH for these cases.

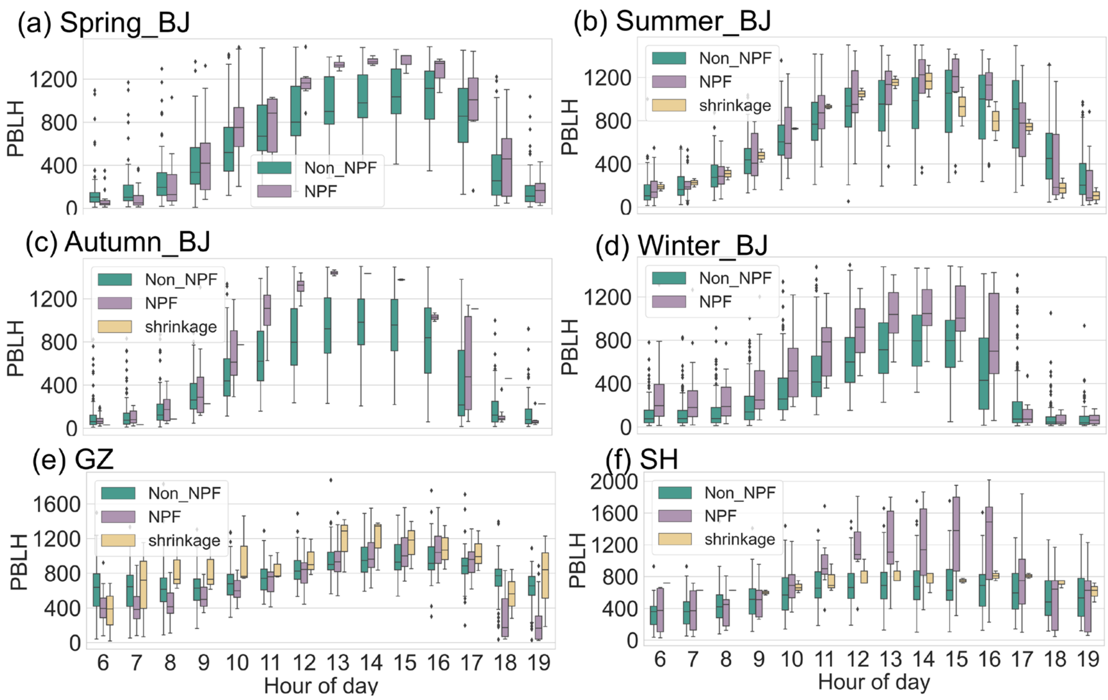

Fig. R2. Diurnal variations of planetary boundary layer height (PBLH) on NPF days (purple), Non_NPF days (green), and shrinkage events (yellow) in (a–d) different seasons in Beijing, (e) Guangzhou, and (f) Shanghai.

Reviewer Comment:

Problematic interpretations and vague statements. There are too many, and I just list few here:

Line 57. The impact of PBLH on NPF is complex and significant, involving … Please specify in which way PBLH can influence particle growth mechanisms.

Response:

We appreciate this suggestion and have revised the manuscript accordingly for clarity. As the boundary layer begins to rise, atmospheric particles become well mixed and diluted, leading to a decrease in aerosol concentrations and a cleaner CS, which increases the likelihood of NPF events (Blanco-Alegre et al., 2022; Nilsson et al., 2001). Additionally, aircraft measurements have shown that the development of the PBL and associated vertical mixing can promote the burst of ultrafine particles within the residual layer (Platis et al., 2015), and are highly correlated with the occurrence of NPF events (Blanco-Alegre et al., 2022). These details are now explicitly discussed in Section 2.3 of the revised manuscript.

Line 100. HYSPLIT model does not consider emissions.

Response:

Thank you for pointing this out. We fully agree that the HYSPLIT model does not explicitly simulate emissions or chemical production processes. In our study, HYSPLIT was used solely to calculate backward air-mass trajectories, with the purpose of identifying the transport pathways and potential source regions of the air masses arriving at the measurement sites. Our interpretation is therefore based on trajectory residence time statistics (PSCF/CWT) rather than on emission modeling.

We also appreciate the reviewer’s comment on this point and acknowledge that there were indeed some errors in our original manuscript. Specifically, we mistakenly interpreted the regions passed by air masses as pollution source areas, which is a serious misrepresentation. We have corrected this in the revised version. To improve the analysis, we divided each NPF event into three phases and examined the backward trajectories separately for each phase.

Based on 48-hour backward trajectories, we calculated the trajectory residence time for nucleation-mode (Nuc, 11–25 nm), Aitken-mode (25–100 nm), and coarse-mode (>100 nm) particles at the three sites—Beijing, Guangzhou, and Shanghai—and obtained their spatial probability distributions. In all three cities, the high-probability regions for Nuc-mode particles were associated with air masses originating from relatively clean upwind areas. For the Beijing site, air masses from the northwest and north typically carried lower CS levels, favoring SO_2 oxidation to H_2SO_4 and subsequent nucleation. In Guangzhou, air masses predominantly came from the south and southwest, while for Shanghai, high-probability regions were mainly located in inland areas to the west.

The Aitken mode during the selected time periods likely represents particles that have grown from the Nuc mode during transport, reflecting the aging and growth of newly formed particles. Coarse-mode particles mainly reflect regional resuspension and the influence of aged local background aerosols. The coarse mode is more representative of background particle concentrations and locally emitted large particles.

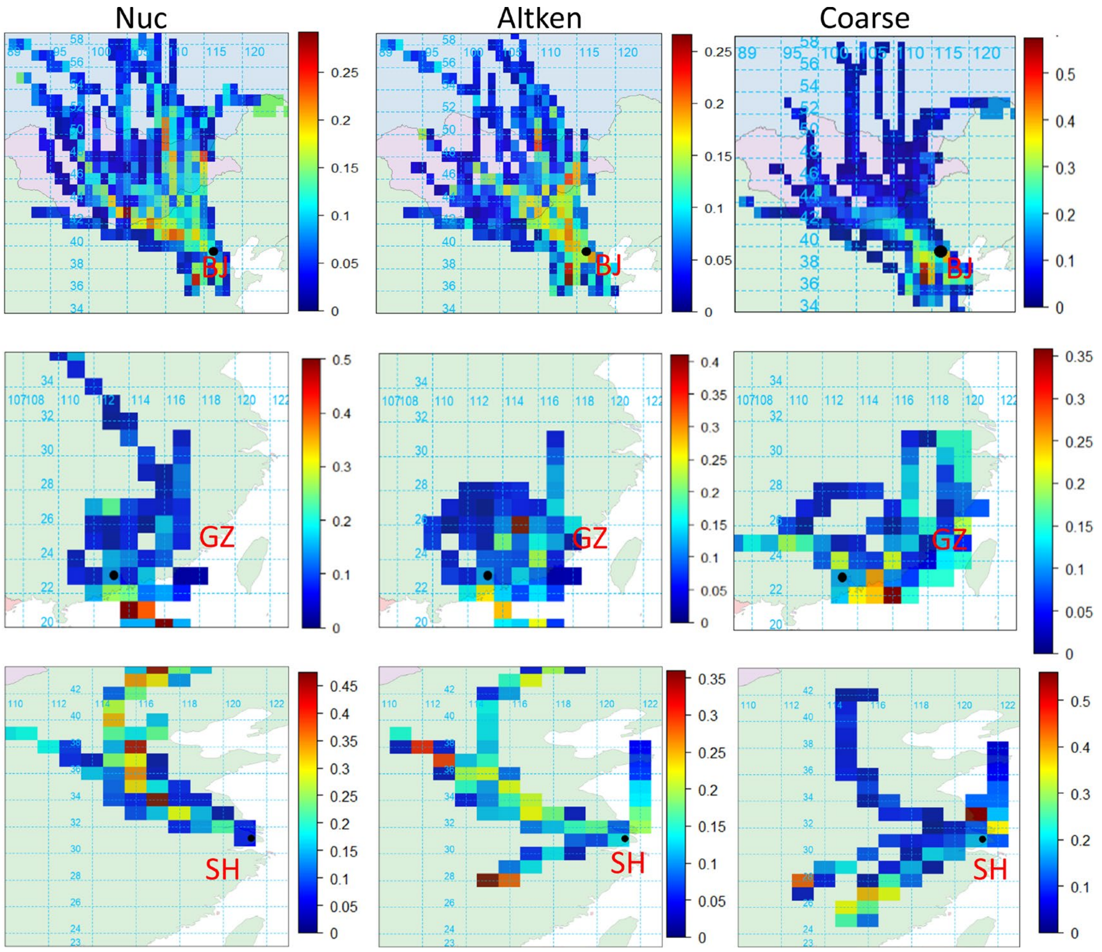

Fig. R3. The 48h backward trajectory by using PSCF, and the map of NPF event contribution levels when particle size is below 100nm in BJ, GZ, and SH during NPF days.

Line 136. I don't understand why primary particles can facilitate new particle growth?

Response:

We apologize for the ambiguity in the original statement. Our intention was to discuss the role of pre-existing ultrafine particles (UFPs) as potential condensation surfaces. Background PNC and CS are related but not equivalent. CS is governed mainly by the surface area of the pre-existing particles, particularly the accumulation mode. In many urban observations, low particle number concentration often coincides with low surface area and therefore low CS, but exceptions occur when the particle population is dominated either by numerous ultrafine particles (high N but low CS) or by a small number of large accumulation-mode particles (low

N but high CS) (Chu et al., 2019; Zhou et al., 2020). This has now been corrected in the manuscript, and the relevant sentence has been rephrased for clarity.

Fig.4, Fig.5, and relevant discussion. The classification of Type I and Type II merely based on their starting time. But this can be easily explained by the dial pattern of nucleating precursors (H2SO4 and others). This can be more relevant to the seasonal change in solar radiation, which just coincides with the evolution of PBLH.

Response:

We appreciate this valuable observation and agree that seasonal variation in solar radiation plays an essential role in regulating the production of nucleating precursors such as H_2SO_4. However, our classification of Type I and Type II events is based on the relative timing of NPF initiation with respect to PBLH growth onset, not solely the time of day. In Type I events, NPF begins almost simultaneously with the start of PBLH development, while in Type II events, a distinct delay is observed between the PBLH rise and the nucleation onset.

In response to the reviewer's comment, we analyzed the variations in SO_2 and also calculated Proxy[H_2SO_4] concentrations. As shown in Figure R4, we selected two representative cases from each event type. Under similar meteorological conditions (e.g., temperature and RH), differences in PBLH evolution led to distinct timings in the onset of NPF.

In the Type 1 case, the onset of the NPF event and the rapid growth of nucleation-mode particles (11–25 nm) were observed near the surface when the convective boundary layer had just begun to develop and PBLH remained relatively low. At this stage, turbulent mixing was only starting to intensify, and the occurrence of NPF was more directly associated with local near-surface radiation conditions and the gradual accumulation of gaseous precursors (e.g., H_2SO_4). In such cases, Proxy[H_2SO_4] typically began to rise only after the growth of 10 nm particles was observed, suggesting that the initial boundary layer development and the concurrent enhancement of local photochemical production jointly triggered the NPF event.

In contrast, the Type 2 case showed that a clear NPF event at the surface only occurred after the PBLH had increased beyond a certain threshold (approximately 800 m). This indicates that in such cases, the triggering of NPF is more dependent on a well-developed mixed layer and strong vertical mixing. Proxy[H_2SO_4] in these cases often reached high levels—even approaching their peak—before the onset of NPF at the surface. However, 10 nm particle bursts were not observed until the PBLH rapidly increased and sufficient mixing between the residual layer or upper-level air and the surface took place, leading to the onset of the NPF event.

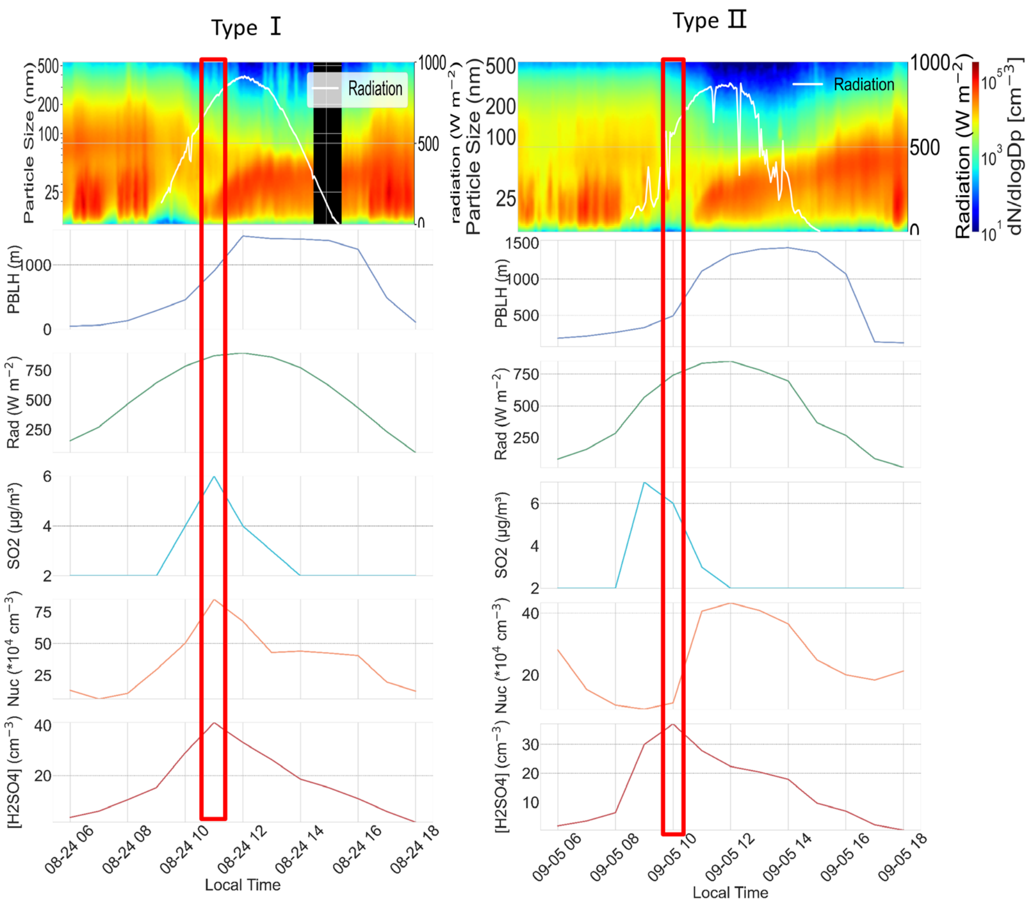

Fig. R4 The diurnal evolution of PBLH, radiation, SO_2, $PNC_{nu}c$, and proxy [H_2SO_4] in Type 1 events, (24 August, 2017), and in Type2 (5 September 2017)

Line 383-384. I question the interpretation of PSCF results. It is well understood that in Beijing, high nucleation-mode particles were often associated with northerly wind, because the clean air with low CS favors NPF. Mongolia should not be considered as a pollution source of nucleation-mode particles.

Response:

Thank you for raising this important point. We fully agree that the northern trajectories, including those from Mongolia, are not necessarily indicative of pollution sources but rather represent air masses with lower CS levels that create favorable conditions for NPF. In the revised discussion of Figure R6, we have rephrased the interpretation to clarify that these regions are associated with favorable formation conditions rather than emission sources. The term "pollution source" has been removed to avoid misunderstanding.

Reviewer Comment:

Line 42, the citation of Chan et al., 2020 is not appropriate, as this paper is mostly about data inversion of PSM.

Response:

We thank the reviewer for this helpful comment. We agree that Chan et al., (2020) mainly focuses on PSM data inversion techniques, and thus it is not the most appropriate reference for supporting the general statement that "many methods have been used to identify the growth characteristics and development mechanisms of NPF."

Reviewer Comment:

Line 51, the citation of Shengjie et al., 2001 seems to be problematic.

Response:

We thank the reviewer for pointing out this issue. We have identified and corrected the citation errors accordingly. The revised statement is as follows:

Shengjie, N., Chengchang, Z., and Jiming, S.: Observational Researches on the Size Distribution of Sand Aerosol Particles in the Helan Mountain Area, Transactions of Atmospheric Sciences, 25, 243-252, 2001.

Reviewer Comment:

Line 53, “it found that a sluggish GR and the presence of pristine background aerosols…” I guess the author meant to say “pre-existing background aerosols”

Response:

We appreciate the reviewer’s correction. The phrase “pristine background aerosols” was incorrectly used, and we have revised it to “pre-existing background aerosols” to better reflect the intended meaning.

Reviewer Comment:

Line 61, “relative humidity”

Response:

We thank the reviewer for pointing this out. The misspelling was due to our oversight, and we have corrected it in the revised manuscript.

Reviewer Comment:

Line 107-110. There is no Chapter in the manuscript.

Response:

Thank you for pointing out the misuse of the term "Chapter" in the manuscript (Lines 107–110).

We agree that this terminology is not appropriate in the context of a scientific article. We have revised all relevant instances to use "Section" instead of "Chapter" to better align with the standard structure of journal articles. We appreciate your careful reading and will ensure consistency in terminology throughout the revised manuscript.

Reviewer Comment:

Line 312. “catalyst” is not properly used in this context.

Response:

We thank the reviewer for this important comment. We agree that the term “catalyst” was not used appropriately in this context. Our original intention was to describe the role of turbulent mixing or boundary layer development in facilitating the vertical exchange of air masses and precursor gases during NPF events, rather than implying a chemical catalytic process.

To address this, we have revised the sentence in Line 312 to remove the term “catalyst” and rephrased it as follows:

The rapid growth of the boundary layer can enhance the upward transport of low-level precursors and the downward entrainment of residual-layer compounds, thereby promoting the occurrence of NPF events (Nilsson et al., 2001).

This revision more accurately reflects the physical mechanism without misusing chemical terminology. We appreciate the reviewer’s careful observation, which helped us improve the clarity and precision of the manuscript.

Reviewer Comment:

Line 380. Fig.8 should be Fig.7

Response:

We acknowledge the incorrect figure reference. The sentence previously referred to "Fig. 8", which was a mistake. We have corrected it to "Fig. 7" in the revised manuscript.

Reviewer Comment:

Line 420-421. Grammatic error

Response:

We have reviewed the sentence and revised it to correct the grammatical error. The revised version reads:

“We identified two three distinct mechanisms of NPF initiation: Type Ⅰ, and Type Ⅱ, and shrinkage.”

Reviewer Comment:

Line 424-425. Grammatic error

Response:

Lines 424–425: This sentence has also been revised for grammatical clarity. The revised version now reads:

“Correlation analyses highlight that the boundary layer plays a dominant role in triggering NPF, particularly at the Shanghai site where the correlation reaches 0.99.”

Blanco-Alegre, C., Calvo, A. I., Alonso-Blanco, E., Castro, A., Oduber, F., and Fraile, R.: Evolution of size-segregated aerosol concentration in NW Spain: A two-step classification to identify new particle formation events, J Environ Manage, 304, 114232, 10.1016/j.jenvman.2021.114232, 2022.

Chan, T., Cai, R., Ahonen, L. R., Liu, Y., Zhou, Y., Vanhanen, J., Dada, L., Chao, Y., Liu, Y., Wang, L., Kulmala, M., and Kangasluoma, J.: Assessment of particle size magnifier inversion methods to obtain the particle size distribution from atmospheric measurements, Atmospheric Measurement Techniques, 13, 4885-4898, 10.5194/amt-13-4885-2020, 2020.

Chu, B., Kerminen, V.-M., Bianchi, F., Yan, C., Petäjä, T., and Kulmala, M.: Atmospheric new particle formation in China, Atmospheric Chemistry and Physics, 19, 115-138, 10.5194/acp-19-115-2019, 2019.

Mengqi, L., Xiangao, X., and Jinqiang, Z.: A value-added surface shortwave radiation dataset at Xianghe.csv, Science Data Bank [dataset], doi:10.57760/sciencedb.02058, 2022.

Nilsson, E. D., Rannik, Ü., Kulmala, M., Buzorius, G., and O'dowd, C. D.: Effects of continental boundary layer evolution, convection, turbulence and entrainment, on aerosol formation, Tellus B: Chemical and Physical Meteorology, 10.3402/tellusb.v53i4.16617, 2001.

Platis, A., Altstädter, B., Wehner, B., Wildmann, N., Lampert, A., Hermann, M., Birmili, W., and Bange, J.: An Observational Case Study on the Influence of Atmospheric Boundary-Layer Dynamics on New Particle Formation, Boundary-Layer Meteorology, 158, 67-92, 10.1007/s10546-015-0084-y, 2015.

Sun, Y. L., Wang, Z. F., Du, W., Zhang, Q., Wang, Q. Q., Fu, P. Q., Pan, X. L., Li, J., Jayne, J., and Worsnop, D. R.: Long-term real-time measurements of aerosol particle composition in Beijing, China: seasonal variations, meteorological effects, and source analysis, Atmospheric Chemistry and Physics, 15, 10149-10165, 10.5194/acp-15-10149-2015, 2015.

Zhou, Y., Dada, L., Liu, Y., Fu, Y., Kangasluoma, J., Chan, T., Yan, C., Chu, B., Daellenbach,

K. R., Bianchi, F., Kokkonen, T. V., Liu, Y., Kujansuu, J., Kerminen, V.-M., Petäjä, T., Wang, L., Jiang, J., and Kulmala, M.: Variation of size-segregated particle number concentrations in wintertime Beijing, Atmospheric Chemistry and Physics, 20, 1201-1216, 10.5194/acp-20-1201-2020, 2020.