We are very grateful to the evaluations from the reviewers, which have allowed us to clarify and improve the manuscript. Below we addressed the reviewer comments, with the reviewer comments in black and our response in blue. The changes in the revised manuscript are indicated in italics and line number that we refer to is the tracked version (All Markup) of the revised manuscript.

Reply for the referee #1 - Mao et al.

General comments: The default BHN nucleation scheme in CMAQ model (version 5.3.2) underestimates particle number concentrations. To address this, Mao et al. integrated and tested multiple parameterizations (BHN, THN, IMN, DMA, SumBTD, and SumID) into CMAQ v5.3.2. Among these, the SumID scheme (IMN+DMA) proved most effective, substantially improving the simulation of number concentrations and NPF events in Beijing and Nanjing. The enhanced model reduces the underestimation by 36–84% compared to observations, in contrast to the default BHN scheme, which misses 70–78%. Results highlight the dominant role of IMN (~56% in Beijing, ~28% in Nanjing) and the significant contribution of DMA (~28–29%). The IMN shows a strong diurnal variability, with higher concentrations during the daytime and lower at night, while DMA specifically influences the morning and evening peaks. Overall, the study demonstrates that incorporating multiple nucleation pathways improves the ability of the model (CMAQ) to represent NPF processes and quantify the contributions of individual nucleation mechanisms. Furthermore, the findings suggest that nucleation mechanisms vary between regions, emphasizing the importance of multiple nucleation parametrization schemes which this paper demonstrates through the performed case studies for the two different regions.

The study outlines a comprehensive overview of different nucleation parametrization schemes, underscoring the most effective which contribute substantially to the model advancements and future applications. Overall, the paper is clearly written and logically organized, however, some modest technical improvements could further enhance the readability. I have some comments/suggestions/questions, mostly for the clarification, that should be addressed before publication.

General Response: We greatly appreciate the referee for their time and efforts devoted to the review of our submission. We realize that most of the comments arise from the inaccuracies or lack of coherence in the content description. We will present these details in the following responses.

Specific comments and responses:

General remark: several acronyms are used throughout the manuscript, but not all are defined upon first use. For the clarity and better readability, I recommend that the authors ensure all acronyms are spelled out when they first appear in the text.

Response: We apologize for the oversight regarding the annotation of abbreviations. We have added annotation in Line 32, 88, 202, 276.

Line 32: "*We then implement Ternary Homogeneous Nucleation (THN), Ion Mediated Nucleation (IMN), and sulfuric acid-dimethylamine (DMA) nucleation parameterization schemes into the CMAQ model.*"

Line 88: "*Based on this result, the Weather Research and Forecasting model coupled with chemistry (WRF-chem), established the same nucleation parameterization schemes, proves that HIO_3 nucleation is the main nucleated way in the future (Ning et al., 2024).*"

Line 202: "*The current emission inventory only has mass emissions of $PM_{2.5}$ ($PM < 2.5$ μm) and $PM_{2.5-10}$ (2.5 μm $< PM < 10$ μm) and does not contain emission information on the particle number emissions.*"

Line 276: "*Figure 2 shows the predicted and observed number concentrations in PM_1 ($PM < 1.0$ μm), Aitken-mode and Accumulation-mode during the whole sampling episodes.*"

In your results you have shown the comparison between different nucleation schemes for Aitken mode simulations highlighting the relatively high performance by IMN, SumBTD and SumID. However, the manuscript does not provide reasoning for why these schemes outperform the default BHN parametrization especially for the Aitken mode simulations. I suggest to discuss it briefly as it would help readers to better understand the difference in model performance.

Response: Thank you very much for the helpful suggestion. In response to your comments, we have provided additional clarification in line 310: "*Current research indicates that the involvement of amine gases (e.g. NH_3, dimethylamine) (Kirkby et al., 2023; Ning et al., 2024; Feng et al., 2025) and atmospheric ions (Yu et al., 2018; Yu et al., 2020a) can promote the NPF. The main reason for the underestimation of BHN is that this nucleation scheme only considers H_2SO_4 and H_2O, while SumID takes H_2SO_4, amine gases and ions into account simultaneously.*".

Line 329: In the statement, "fine particles are rapidly removed due to condensation and coagulation", the term "removed" is not accurate. In aerosol dynamics, "removal" typically refers to dry/wet deposition or scavenging. Microphysical processes such as condensation and coagulation instead contribute to particle transformation rather than removal.

Response: Thank you for your suggestion of the above details. We have revised the sentence in Line 366 to "*However, the NPF-related nanoparticles undergo rapid transformation through condensation and coagulation when interacting with high concentrations of particles from emissions with elevated ratios. This leaves no opportunity for the nanoparticles to grow over 10 nm (Wu et al., 2007; Guo et al., 2014; Wang et al., 2016; Peng et al., 2021).*".

If I understand correctly (otherwise please clarify), there are some existing studies where CTMs with detailed aerosol microphysics have been applied (for e.g. PMCAMx-UF; doi:10.5194/acp-12-8663-2012). It may be helpful to cite examples of models that do not include aerosol dynamical processes (such as condensation sinks, growth via coagulation etc.), as well as the few that do. This would provide a more comprehensive overview and further highlight the importance of your study.

Response: Thank you for your valuable suggestions. We have added discussions and references in the discussion section in Line 429 to "*Some models have demonstrated that considering the microphysical processes of aerosols can significantly improve the accuracy of PNC simulations (Fountoukis et al., 2012; Zhao et al., 2024). Under high condensation sink, larger particles preferentially condense available vapors, thereby inhibiting the growth of smaller particles. Consequently, the small particles are eventually coagulated by the larger particles, thereby modifying the particle size distribution of the aerosols. But most of the CTMs don't have a relationship between condensation sink, growth rate, and new particle production module. Therefore, this can lead to an overprediction of PNCs under high condensation sink scenarios.*".

Are you certain that your results are representative of all of China? Considering the regional variability mentioned several times in the manuscript, and given that the analysis was performed only for Beijing and Nanjing, I recommend clarifying this point or keep it consistent throughout the manuscript to avoid ambiguity.

Response: We apologize for the overstated conclusions, and have added the limitations of this work. We have changed "China" to "eastern cities" in Line 42, 47, 439.

Line 42: "*This study enhances the model's capability to accurately simulate NPF events and underscores the significant influence of IMN and DMA nucleation on PNC in eastern cities.*".

Line 47: "*CMAQ model incorporated with IMN and DMA nucleation schemes significantly improves its modeling accuracy for particle number concentrations in eastern cities.*".

Line 471: "*Overall, this study validates the enhanced model's capability to accurately simulate NPF events and underscores the significant influence of IMN and DMA nucleation pathways on particle number concentrations in eastern cities.*".

Line 69-70: The sentence states that 'multiple nucleation theories are interrelated and nucleate synergistically in some cases.' This is an interesting point and if I understand correctly, this concept is also considered in your study, but the statement lacks detail. I recommend either elaborating briefly on which cases you are referring to or adding relevant references to support the statement.

Response: Thank you for your valuable suggestions. In this study, we have indeed taken this concept into account in the determination of nucleation parameters. We have added statement in Line 74 and Line 220.

Line 74: "*For example, the IMN theory takes into account BHN, as well as the synergistic effects between THN and ions (Yu et al., 2018; Yu et al., 2020a).*".

Line 219: "*The rationale behind these combinations lies in the different nucleation theories. Since BHN represents the interaction between H_2SO_4 and H_2O, it is a fundamental theory in nucleation mechanisms that exists in most environments (Sipilä et al., 2010). THN represents the involvement of H_2SO_4, H_2O, and NH_3 in nucleation, used to explain the higher nucleation rates in the atmosphere*

(Merikanto et al., 2007). Therefore, the binary and ternary nucleation scenarios are interconnected. But IMN nucleation accounts for the synergistic interactions among BHN, THN, and ions (Yu et al., 2018; Yu et al., 2020a). Consequently, this nucleation mechanism cannot be integrated with BHN and THN scenarios.".

Line 102: What do you mean by the term "episodes"? I recommend defining or specifying these episodes more clearly to avoid ambiguity and improve readability.

Response: We have changed "episodes" to "NPF periods" and revised sentence in Line 116 to "*In this study, we first evaluate the performance of the default CMAQ v5.3.2 model on predicting PNC in Beijing and Nanjing during two NPF periods, and then implement three additional nucleation parameterization schemes into the model, aiming to improve CMAQ's accuracy and to better understand the key nucleation pathways in different urban regions of China.*".

Line 160: The sentence lists the configuration mechanisms used for the gas-phase and aerosol modules but does not provide any details or references in this context for the readers. I suggest briefly elaborating on these mechanisms in one sentence to improve clarity and context.

Response: Thank you for your valuable suggestions. We have added statement in Line 179: "*These settings are currently the best choice for CMAQ to simulate precursors and aerosol chemistry (Li et al., 2022).*".

Line 53 (minor language/grammar suggestions for clarity): The sentence is a bit unclear. For example, the words like "that" and "and" seems to be misplaced here. I suggest revising it to either "in which" or "where" and "that grow into particles…", for the improved readability.

Response: Thank you for your suggestion. We have revised the sentence in Line 56 to "*New particle formation (NPF), a process in which gaseous vapors in the atmosphere form critical molecular clusters that grow into particles (Zhang et al., 2012; Wang et al., 2017), is a significant source of PNC, contributing ~30% of PNC at the surface and over 90% in the upper troposphere (Yu et al., 2020b; Zhao et al., 2024).*".

Line 99: Do you mean "number concentration"?

Response: We apologize for the incorrect writing. We have revised the sentence in Line 108 to "*Currently, there are no studies evaluating the effects of number concentration simulations of IMN and DMA in China.*".

Line 141 (minor language/grammar suggestions for clarity): The sentence looks incomplete. I suggest to please check and rephrase this statement.

Response: We have revised the sentence in Line 156 to "*This look-up table encompasses the considerations of BHN, Binary IMN (BIMN), THN, and Ternary IMN (TIMN).*".

Line 148: Do you mean “thermodynamic theory”?

Response: We have revised the sentence in Line 163 to “*In earlier classical binary and ternary nucleation parameterizations, RH is indispensable, which are based on classical kinetics and thermodynamic theory.*”.

Figure 1 (minor suggestion): since the unit (cm-3) is common across all figures (y-axis), I suggest incorporating it directly into the y-axis label and possibly renaming the label for clarity, if possible.

Response: Thank you for your valuable suggestions. We have revised the labels in Figure 1.

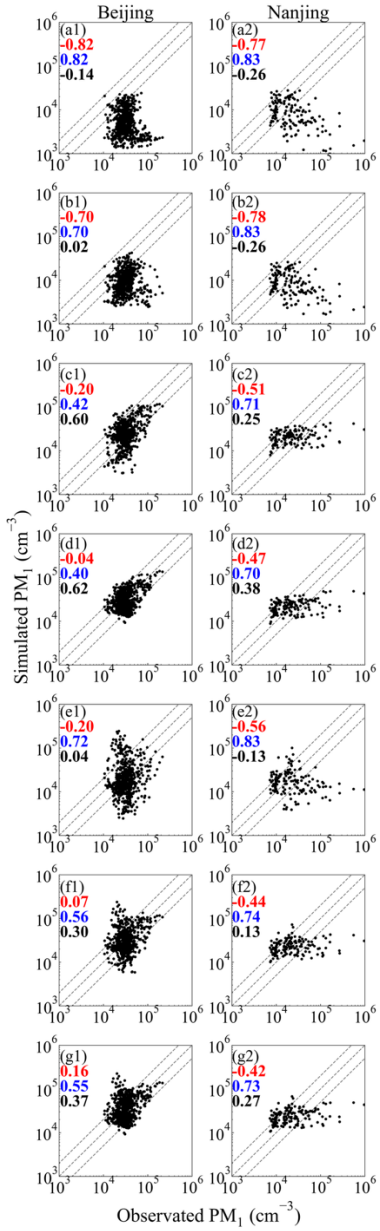

Figure 1. Model performance of PM₁ number concentration in (1) Beijing and (2) Nanjing. Each point corresponds to a 1-h average value. (a) None scenario. (b) BHN scenario. (c) THN scenario. (d) IMN scenario. (e) DMA scenario. (f) SumBTD scenario. (g) SumID scenario. The 1:1, 1:2, and 2:1 line are

shown. NMB (red font): normalized mean bias; NME (blue font): normalized mean error; R (black font): correlation coefficient.

Figure 2 (minor suggestion): if possible, please increase the font size and add subplot titles (e.g., "Beijing" and "Nanjing") for consistency with Fig. 1.

Response: Thank you for your valuable suggestions. We have revised the font size and titles in Figure 2.

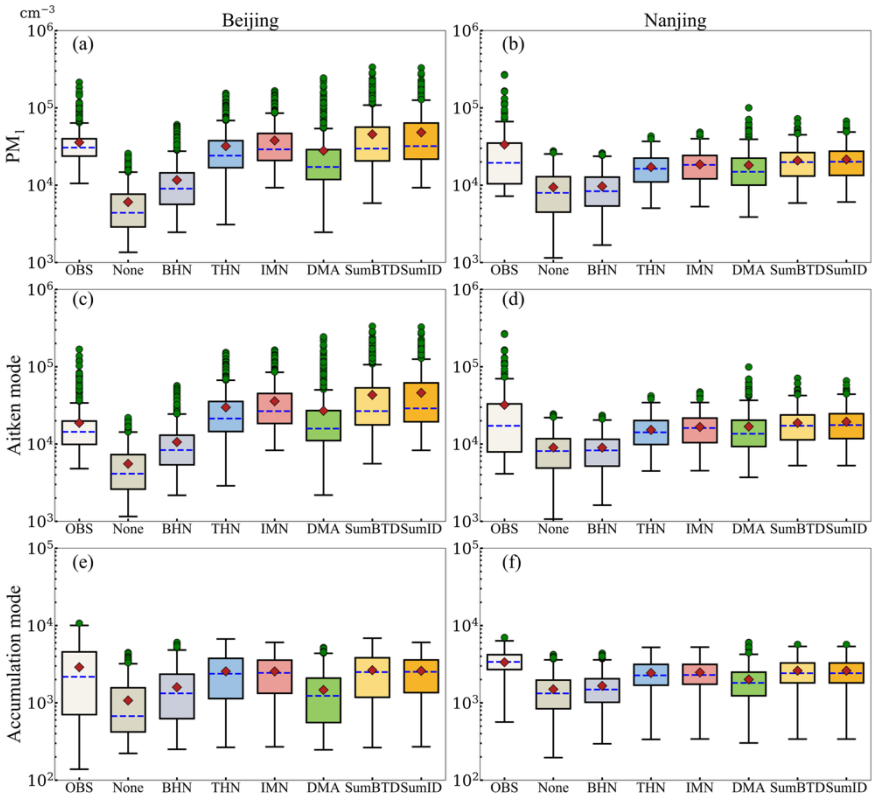

Figure 2. Comparison of (a), (b) PM_1, (c), (d) Aitken-mode, and (e), (f) Accumulation-mode number concentration in seven scenarios with observations during NPF events in (a, c, e) Beijing and (b, d, f) Nanjing. The dashed blue line is the median value, and red prism is the mean value.

Line 301-303 and Figure 3: In the following lines, you describe morning and evening peaks as well as daytime values. If I understand correctly, these refer to Figure 3. However, the figure is somewhat difficult to follow. First, the color representation for 'None,' 'IMN,' and 'DMA' is not clearly distinguishable, please consider changing the colors, making them darker, or increasing the line width. Second, I suggest adding background shading to highlight daytime and nighttime periods, which would make the figure easier to interpret.

Response: Thank you for your valuable suggestions. We have revised the colors, line width and shading in Figure 3.

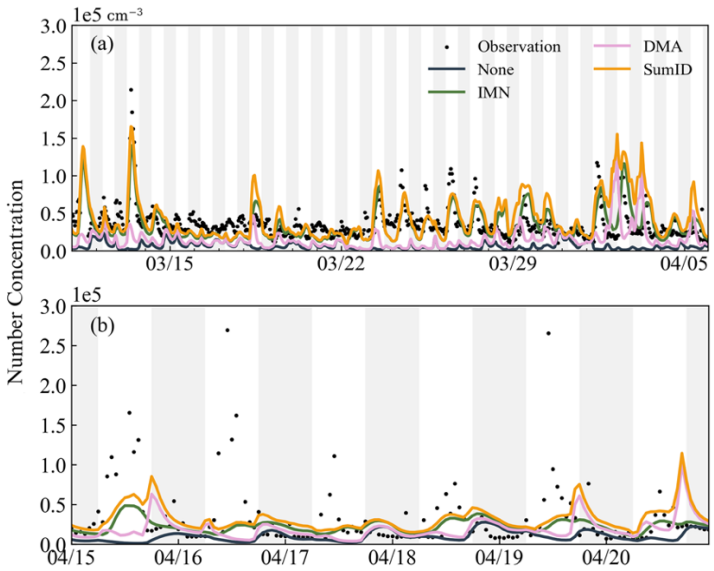

Figure 3. Observation and four schemes of None, IMN, DMA and SumID in hourly particle number concentrations (cm^{-3}) in (a) Beijing and (b) Nanjing. Background shading represents nighttime periods.

Line 306 (minor language/grammar suggestions for clarity): The sentence beginning with "Among of them,…" is not grammatically correct. I suggest changing it to "Among them..".

Response: We have revised the sentence in Line 342 to "*Among them, the events on the 16th and 17th are not captured by the model (Figure 3b).*".

Line 322: please specify the month also to avoid ambiguity.

Response: We have revised the sentence in Line 360 to "*Due to the inability to capture two NPF events on the April 16th and 17th in Nanjing, these events are excluded from the analysis.*".

Supplement figure S3 (minor suggestion): since this figure presents an important finding of your study, I recommend increasing the font size for better readability, particularly on the x-axis, which is difficult to see.

Response: Thank you for your valuable suggestions. We have revised the font size in Figure S3.

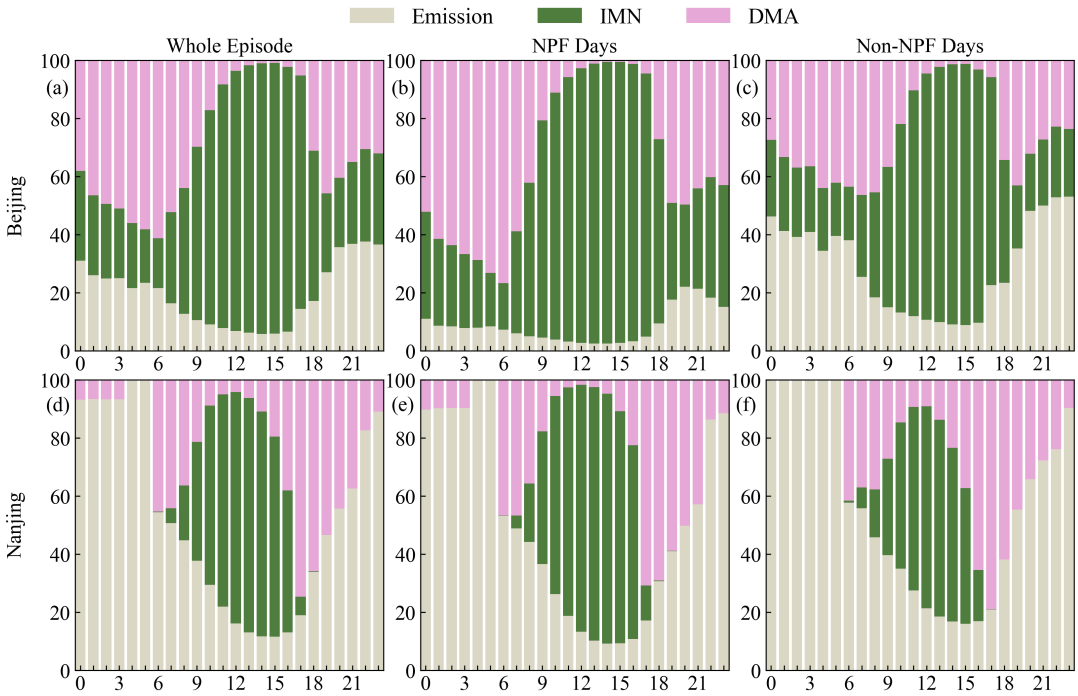

Figure S3. The average diel variation of the hourly contributions (%) in whole episode, NPF days and non-NPF days.

Line 347: In this sentence you have mentioned “three types of days”, which is then described in the supplement figure. I suggest elaborating on this directly in the text rather than only referring to the supplementary figure, so that readers can understand it without needing to cross-reference.

Response: Thank you for your valuable suggestions. We have added annotation in Line 389: “*In Figure S3, IMN contributes a high number concentration in the daytime with a notable diurnal variation both in Beijing and Nanjing across entire periods, NPF days, and non-NPF days.*”.

Figure 5: same as for Figure 3. Please add the background shading to highlight day and night time periods and increase the font size, if possible. Additionally, I suggest incorporating the unit of the nucleation rate (cm-3 s-1) directly into the y-axis label.

Response: Thank you for your valuable suggestions. We have revised the font size and label in Figure 5.

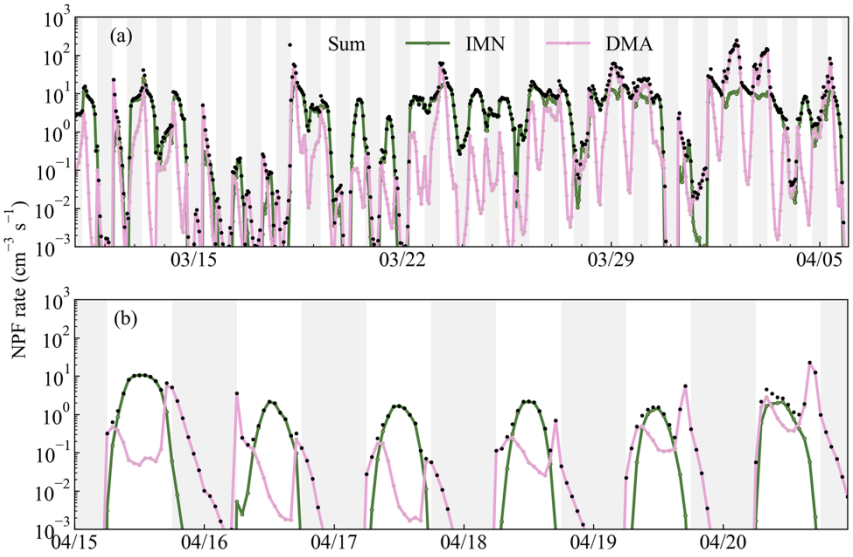

Figure 5. NPF hourly particle nucleation rate (cm^{-3} s^{-1}) of two nucleation parameterizations within SumID nucleation parameterization in (a) Beijing and (b) Nanjing. Background shading represents nighttime periods.

Line 388: The sentence appears incomplete "…subsequently lost from the system." Please explicitly mention the processes responsible for the particle removal from the system to clarify the statement.

Response: We have revised the sentence in Line 432 to "*Consequently, the small particles are eventually coagulated by the larger particles, thereby modifying the particle size distribution of the aerosols.*".

Line 398: Do you mean "multiple nucleation schemes"?

Response: We apologize for the inaccurate writing. We have revised the sentence in Line 454 to "*After integrating multiple nucleation schemes into the aerosol module of CMAQ v5.3.2, including BHN, THN, IMN, DMA, SumBTD (BHN+THN+DMA), and SumID (IMN+DMA), the SumID nucleation scheme was identified as the optimal approach.*".

Line 415-417: This statement repeats the content from lines 403-405. Please consider removing or rephrasing to avoid redundancy.

Response: We apologize for the repeated writing. We have removed sentence in Lines 403-405 in preprint version.

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Reply for the referee #2 - Mao et al.

General comments: Particle number concentrations, as computed by the available parametrization scheme for nucleation within the CMAQ model, did not agree well with observations (in China), and the authors have incorporated few other schemes and conducted simulations. Along with few individual schemes, numerical experiments combining few schemes together, have also been carried out. The Paper suggests enhanced capability of the model with the implemented schemes.

Overall manuscript is a significant advancement and the discussions are clear. The manuscript is recommended for publication in GMD. The following comments are provided for authors to consider during the revision process.

General Response: We greatly appreciate the referee for their time and efforts devoted to the review of our submission. We realize that most of the comments arise from the unclear in the content description. We will present these details in the following responses.

Specific comments and responses:

l.81: "WRF-Chem proves that HIO_3 nucleation is the main way in future". This is not very clear. Review and reframe this discussion.

Response: We apologize for any inaccuracies in the expressions. We have revised the sentence in Line 85 to "*By coupling all the different forms of nucleation parameterization schemes discussed above, the E3SM model indicates that H_2SO_4-dimethylamine nucleation dominates number concentration in 1 km height (Zhao et al., 2024). Based on this result, the Weather Research and Forecasting model coupled with chemistry (WRF-chem), established the same nucleation parameterization schemes, proves that HIO_3 nucleation is the main nucleated way in the future (Ning et al., 2024).*".

l.97-98: The approach SumBTD and SumID are not clear to me. I did not find much description of these approaches. How do you combine several options of nucleation together? How do you decide which of the options should be combined like in case of BTD or in case of ID, instead of other possible combinations?

Response: Thanks for your comment. To clarify this, we have revised the sentence in Line 219 to "*The rationale behind these combinations lies in the different nucleation theories. Since BHN represents the interaction between H_2SO_4 and H_2O, it is a fundamental theory in nucleation mechanisms that exists in most environments (Sipilä et al., 2010). THN represents the involvement of H_2SO_4, H_2O, and NH_3 in nucleation, used to explain the higher nucleation rates in the atmosphere (Merikanto et al., 2007). Therefore, the binary and ternary nucleation scenarios are interconnected. But IMN nucleation accounts for the synergistic interactions among BHN, THN, and ions (Yu et al., 2018; Yu et al., 2020). Consequently, this nucleation mechanism cannot be integrated with BHN and THN scenarios. Given that DMA nucleation, containing H_2SO_4 and dimethylamine, has been identified as the predominant nucleation mechanism in urban environments (Yao et al., 2018; Liu et al., 2021; Wang et al., 2021), this study specifically addresses urban nucleation mechanism. Therefore, the DMA nucleation scheme*

must be incorporated into the combined scenarios. Based on the discussed above, we have retained only two combined scenarios for further analysis.".

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