RE: A point-to-point response to referees' comments

We thank the editor for handling this manuscript and the referees for their helpful and insightful comments. To address the concerns raised by the referees, we have strengthened and clarified the evidence for our main conclusions and improved the writing for better understanding. Point-by-point responses to the comments are given below. The referees' comments are shown in *red italics*. Our responses are given in black. Revised text is shown in blue in the response. Line numbers in the responses correspond to those in the revised manuscript.

Referee #1:

1. This paper describes results from atmospheric nucleation studies in Chinese cities and its technical aspects appear to be at the state of the art in this area. While the manuscript is admirably succinct, it also admirably touches on all (at least most) of the aspects of the important information. Yet there are sentences that are difficult to understand, and it seems that some inferences and conclusions are not well-supported (the first may be causing some of the second.) The main conclusion of the paper is that H_2SO_4-DMA nucleation can explain the observations in these cities. A secondary one is that OOMs are needed to explain growth rates of newly nucleated particles.

Response:

Thanks for these positive and constructive comments. We have improved the writing and rewritten the sentences that were difficult to follow. We hope the revised manuscript clarifies the evidence for our main conclusions (see below).

2. Support for the main conclusion is not well presented. For example, line 165 states that SA-DMA was 'identified ' as the dominate (sic) NPF mechanism. This is not a finding if it is solely based on Figure 1 (correlation and causation are falsely linked.) For example, line 165 states that SA-DMA was 'identified ' as the dominate (sic) NPF mechanism. This is not a finding if it is solely based on Figure 1 (correlation and causation are falsely linked.) The analysis presented in section 3.2 is focused on teasing out SA and temperature dependencies. Should not a dependence on DMA be important for identifying SA-DMA nucleation? Figure S3 is mentioned as supporting the SA-DMA mechanism but on the face of it the $J_{1.4}$ sim.-meas. correlation is not good. Similarly for $J_{1.7}$ (figure 7b). For both of those, it looks like the measurements range over about 1/2 the orders of magnitude that the simulations do: this is not support for the mechanisms in the simulations. Assuming SA dimer is an indicator of SA-DMA nucleation, the best support is Figure 4 but SA_2 scaled is not presented here (confused by the text on lines 220-224.) Figure 5 does present SA_2 scaled instead. Good: it does show a decently strong temperature dependence (would be helpful to have data in Figure 4 colored by temperature also.) But two things are bothersome in this analysis: the above-mentioned dependence on DMA is washed away and details are scant on the reason behind the temperature dependence of the simulations (bond strength, and only for the SA.DMA cluster?)

Response:

We thank the referee for these constructive comments. To better support the main conclusions of this study, we

have revised the manuscript in the following aspects:

(1) We have revised the description and implication of each figure, and removed misleading statements that are not supported by the figures.

(2) We have added a correlation analysis between neutral SA-DMA clusters and the number concentration of sub-3 nm particles to provide support for SA-DMA nucleation.

(3) We have rewritten the discussions on the influence of DMA on nucleation.

(4) We have added statistics of the NPF frequency in different campaigns. The NPF frequency shows a strong negative correlation with temperature, providing support for the significant influence of temperature on atmospheric nucleation.

(5) We have expanded the discussion to better explain the inhibitory effect of temperature on NPF.

(6) We have added results of multiple field studies outside China, and compared them with those in this study, highlighting the main contributions and the global relevance of our work.

With these improvements, we think the support for our inferences and conclusions is clarified. We briefly summarize the main conclusions and their supporting evidence below:

Conclusion #1: SA-DMA nucleation can explain the observations in these cities. Supporting evidence includes:

(1) A good correlation between particle formation rate and [SA_1] was observed in our field measurements. The results for nucleation rate versus [SA_1] indicate highly efficient nucleation mechanisms, consistent with the curves from CLOUD experiments and other atmospheric studies under SA-DMA nucleation (Figure 1);

(2) Neutral clusters composed of SA and DMA were detected, and their signals were strongly correlated with the concentration of sub-3nm particles, demonstrating their involvement in cluster formation (Figure 2 and Figure S3);

(3) Both measured [SA_2] and particle formation rates aligned with theoretical predictions using the SA-DMA nucleation mechanism (Figure 4 and Figure S7);

(4) The dependence of NPF on influencing factors, namely temperature and [DMA], was also consistent with theoretical predictions (Figures 5-6 and Figure S5).

Conclusion #2: OOMs are needed to explain the growth rates of newly nucleated particles. This is supported by the fact that the growth contributed by SA and its clusters cannot explain the initial growth (1.4-1.7 nm), and the improved agreement between measured and predicted $J_{1.7}$ when the contribution of OOMs to particle growth was considered (Figure 7).

In addition to these two main conclusions summarized by the referee, the major implication of this study is given: SA-DMA nucleation can explain atmospheric NPF over a large spatial scale of polluted atmosphere, and temperature is a major cause for the difference in NPF characteristics at different locations. This is supported by:

(1) The sites span a broad geographic range and encompass a variety of urban and suburban environments (Figure S1).

(2) A negative correlation between NPF frequency and ambient temperature (Figure 3c-d).

(3) The dependences of NPF on [SA_2] and particle formation rate (Figure 5-6).

The specific comments are copied below to facilitate our point-by-point response.

2.1. For example, line 165 states that SA-DMA was 'identified ' as the dominate (sic) NPF mechanism. This is not a finding if it is solely based on Figure 1 (correlation and causation are falsely linked.) For example, line 165 states that SA-DMA was 'identified ' as the dominate (sic) NPF mechanism.

Response:

Thanks, we have revised the original statement "the dominate NPF mechanism in our campaigns was identified as SA-DMA" into "the observed nucleation is likely driven by SA and enhanced by strong stabilizing precursors such as DMA. Other nucleation mechanisms are unlikely to dominate under these conditions" (lines 172-173). Here we use the relationship between the particle formation rates and [SA_1] as supporting evidence for nucleation mechanisms. It was insufficient to 'identify' a nucleation mechanism with this evidence. We presented more evidence to support SA-DMA nucleation, and all the evidence has been summarized (lines 158-161):

"SA and DMA can explain the atmospheric nucleation observed at all sites. This finding is supported by the correlation between [SA_1] and particle formation rates, the composition of detected clusters, the alignment of simulated and measured nucleation intensity ([SA_2] and particle formation rates), and the dependences of NPF on temperature and [DMA]. The first two points are addressed in this section, while the remaining evidence will be discussed in Sect. 3.2 and Sect. 3.3."

2.2. The analysis presented in section 3.2 is focused on teasing out SA and temperature dependencies. Should not a dependence on DMA be important for identifying SA-DMA nucleation?

Response:

Thanks, we have added a paragraph on analyzing the role of DMA in NPF (lines 216-227):

"Besides SA, other potential precursors related to NPF in polluted regions including DMA, NH_3 and OOMs (Figure S4). These species are known to enhance SA-driven nucleation (Kirkby et al., 2011; Almeida et al., 2013; Riccobono et al., 2014) and promote particle growth (Tröstl et al., 2016). Increasing [DMA] considerably enhances SA_2 formation under fixed temperature ranges (Figure S5), and this effect diminishes as [DMA] approaches nucleation saturation (Almeida et al., 2013). This phenomenon supports the contribution of DMA to atmospheric nucleation and is consistent with the results from a flow reactor (Jen et al., 2014). In CLOUD studies, the contributions of OOMs and NH_3 to cluster formation were almost negligible in the presence of DMA (Kürten et al., 2018; Xiao et al., 2021). Given that our field measurements exhibited similar [NH_3] and relatively low [OOMs] compared to those in CLOUD experiments (Table 2), it follows that their effects are also minor under our ambient conditions. Despite the potential participation of other precursors, the observation-simulation agreement suggests DMA is a major base that stabilizes SA clusters (Figure S5). However, [DMA] and [NH_3] were not markedly elevated during NPF periods (Figure S4). The likely reason is that the suppression of NPF by high CS masked the enhancing effect of DMA and NH_3, as they were positively correlated with CS (r^2=0.31 and 0.39, respectively, Figure S6)."

2.3. Figure S3 is mentioned as supporting the SA-DMA mechanism but on the face of it the $J_{1.4}$ sim.-meas. correlation is not good. Similarly for $J_{1.7}$ (figure 7b). For both of those, it looks like the measurements range over about 1/2 the orders of magnitude that the simulations do: this is not support for the mechanisms in the simulations.

Response:

Due to the uncertainties in determining the measured *J* from the particle size distributions, and potentially the fact that our theoretical predictions were solely based on the SA-DMA mechanism, which may miss other influencing factors on NPF, we did not expect a good correlation between the measured and predicted *J* in real atmospheric studies. Instead, we focus on the consistency in the order of magnitude of the measured and predicted *J* values. We argue that, considering the measurement uncertainties, this is an acceptable uncertainty range for atmospheric studies. We have added clarification on this point (lines 127-142 in the Supplement):

"There is an acceptable consistency between $J_{1.4,meas}$ and $J_{1.4,sim}$ when observational and model uncertainties are considered (Figure S7). Deviations reported in previous laboratory or field studies were typically within one order of magnitude (Kürten et al., 2018; Cai et al., 2021; Xiao et al., 2021), smaller than those in our study. This discrepancy likely arises from differences in uncertainty ranges. For a single observation, the measurement uncertainty is relatively low (Freshour et al., 2014), and the resulting simulation uncertainty can be controlled within a narrow range. However, our measurement spanned several years and involved multiple sites, may amplifying the overall uncertainty in $J_{1.4}$.

The uncertainty range of $J_{1.4,sim}$ need to be analyzed basically. Overall, the output range spans approximately three orders of magnitude, indicating high model sensitivity to input parameters. Theoretically, particle formation rate is approximately proportional to $[SA_1]^4$ in SA-DMA nucleation, and this relationship is particularly evident under high CS and high [DMA] (Cai et al., 2021). Considering the uncertainty of $[SA_1]$ merely (+100%/-50%, Table S1), the propagated uncertainty of $J_{1.4,sim}$ is estimated to be +1600%/-94%. Since there is no simple algebraic relation between [DMA] and $J_{1.4}$, their quantitative dependence need be inferred empirically. An urban study reflected that $J_{1.4}$ roughly varied in proportion to twice of the change of [DMA] (1-5 pptv) accounting for other influencing factors (Cai et al., 2021). Considering the uncertainty of [DMA] (+150%/-60%) merely, the uncertainty of $J_{1.4,sim}$ is estimated to be +400%/-80%. In short, by superimposing the uncertainty of input values, the overall uncertainty in modeled $J_{1.4}$, resulting from the propagation of measured precursor uncertainties, is quantitatively reasonable."

2.4. Assuming SA dimer is an indicator of SA-DMA nucleation, the best support is Figure 4 but SA_2 scaled is not presented here (confused by the text on lines 220-224.)

Response:

We agree with the referee that Fig. 4 serves as evidence supporting SA-DMA nucleation mechanism, and the discussion regarding $SA_{2,scaled}$ has been deferred to the next paragraph to maintain clarity and prevent readers being confused. Furthermore, an additional purpose of this figure is to evaluate the reliability of the model, thereby supporting the subsequent scaling of relevant parameters. We have revised this paragraph (lines 248-254) as:

"Figure 4 shows that $[SA_2]_{meas}$ is in accordance with $[SA_2]_{sim}$ when considering the uncertainties, indicating SA and DMA could explain the formation of SA_2. To be specific, $[SA_2]_{meas}$ is slightly lower than $[SA_2]_{sim}$ overall. Similar discrepancies between measured and simulated cluster concentrations have also been reported in CLOUD experiments (Kürten et al., 2014). This systematic underestimation is likely attributable to measurement errors, because not all SA_2 is fully detected, as some may dissociate within the mass spectrometer (Alfaouri et al., 2022). The observation-simulation comparison of $J_{1.4}$ is shown in Figure S7, where simulation results fall within acceptable ranges upon uncertainty analysis. The consistency between measured and simulated parameters ($[SA_2]$

and $J_{1.4}$) supports the significance of SA-DMA collision in NPF."

2.5. Figure 5 does present SA_2 scaled instead. Good: it does show a decently strong temperature dependence (would be helpful to have data in Figure 4 colored by temperature also.) But two things are bothersome in this analysis: the above-mentioned dependence on DMA is washed away and details are scant on the reason behind the temperature dependence of the simulations (bond strength, and only for the SA.DMA cluster?)

Response:

For the colored data points, we have revised not only Figure 4 (lines 256-258), but also Figure S7 (lines 190-192 in the Supplement) as:

"

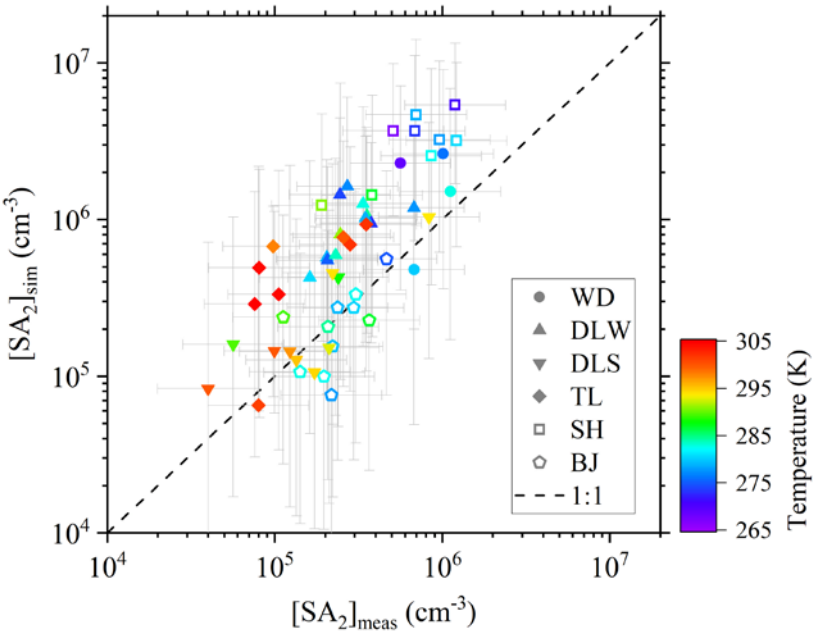

Figure 4: The comparison between $[SA_2]_{meas}$ and $[SA_2]_{sim}$. Horizontal and vertical error bars connected with each symbol indicate the uncertainties of x-axis and y-axis, respectively."

“

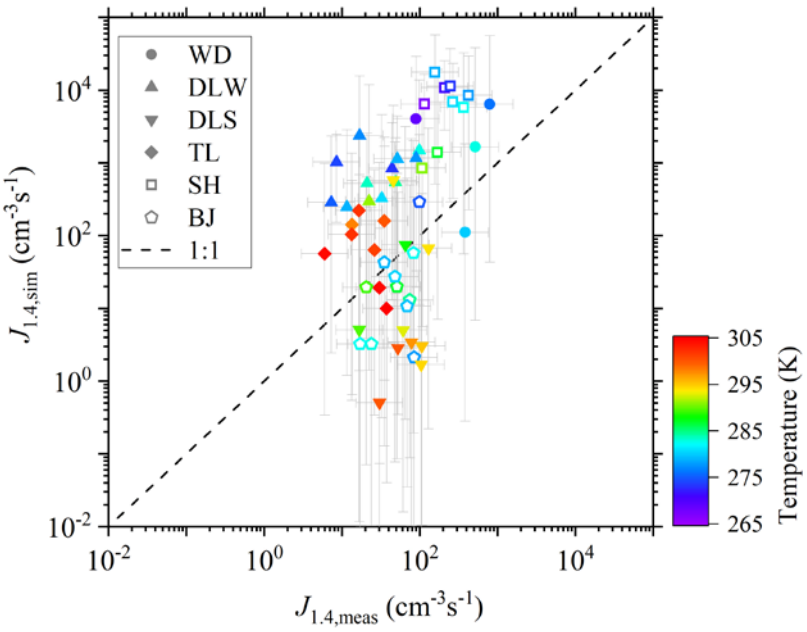

Figure S7: The comparison between $J_{1.4,meas}$ and $J_{1.4,sim}$. Horizontal and vertical error bars connected with each symbol indicate the uncertainties of x-axis and y-axis, respectively.”

For the insufficient analysis of DMA, the response to Comment #2.2 explains this concern. In short, it is undeniable that DMA plays a critical role in the analysis of nucleation mechanisms. We were concerned that readers might overlook the significance of DMA, so we have elaborated on this aspect more clearly.

For the reason behind the temperature dependence of the simulations, we attribute the main cause to the thermal stability of the clusters, and have revised our manuscript as (lines 83-89 in the Supplement):

“Temperature modulates nucleation processes by altering the evaporation rates of clusters, which were derived from collision coefficients and the cluster formation free energy referenced from the literature (Olenius et al., 2017). Among these clusters, SA_1DMA_1 exhibits the most pronounced temperature dependence, and its net formation represents the dominant pathway through which temperature modulates nucleation processes. Reported values for the Gibbs free energy of formation (△G) of SA_1DMA_1 at 298.15 K range from -11.0 to -15.4 kcal mol^{-1} (Olenius et al., 2017; Myllys et al., 2019; Ge et al., 2020; Han et al., 2020). Here, it was set to be -13.5 kcal mol^{-1} at 298.15 K, according to the value from Myllys et al. (2019).”

Moreover, several references to the temperature dependence of simulations were also presented in lines 263-267:

“At the molecular level, it is proposed that SA and DMA form SA_1DMA_1 clusters, which subsequently contributes to SA_2 formation, during nucleation (Olenius et al., 2017; Myllys et al., 2019). While SA_2 with one or two DMA molecules has been demonstrated to already be stable against evaporation (Jen et al., 2014), the formation of SA_1DMA_1 is a temperature-sensitive process that acts as the major rate-limiting step in clustering (Cai et al., 2022b).”

and lines 272-275:

“Theoretically, the evaporation rates of SA trimers and tetramers show limited sensitivity to temperature (Olenius et al., 2017), and their variation can be even neglected in some cluster dynamics models (Cai et al., 2021). Thus,

the observed reduction in larger clusters at elevated temperatures is likely constrained by the formation of SA_2."

In addition to the point-by-point responses above, we have performed a thorough revision of the manuscript, thereby ensuring that the support for main conclusions is well presented.

3. Figure 2 also gives good indirect support to the (or a) SA-DMA mechanism but can the SA_3 and SA_4 signals be correlated to neutral cluster concentrations? They are the ones with DMA in them! This would be support of a more direct nature. Also, this figure needs a bit more explanation. What is/are S-O ions? Symbol size meaning diameter or area? Since the plots are relatively clean, a few could be tagged with logSignal values.

Response:

Regarding the first comment, we are not sure about what the referee specifically meant by "neutral cluster concentrations". We have interpreted it as "the number concentration of sub-3 nm particles" and have plotted a new figure (Figure S3), which showed good correlation between this quantity and both SA_3 and SA_4. The results indicated that these neutral clusters contributed to NPF. We have revised our manuscript as (line 168-171 in the Supplement):

"

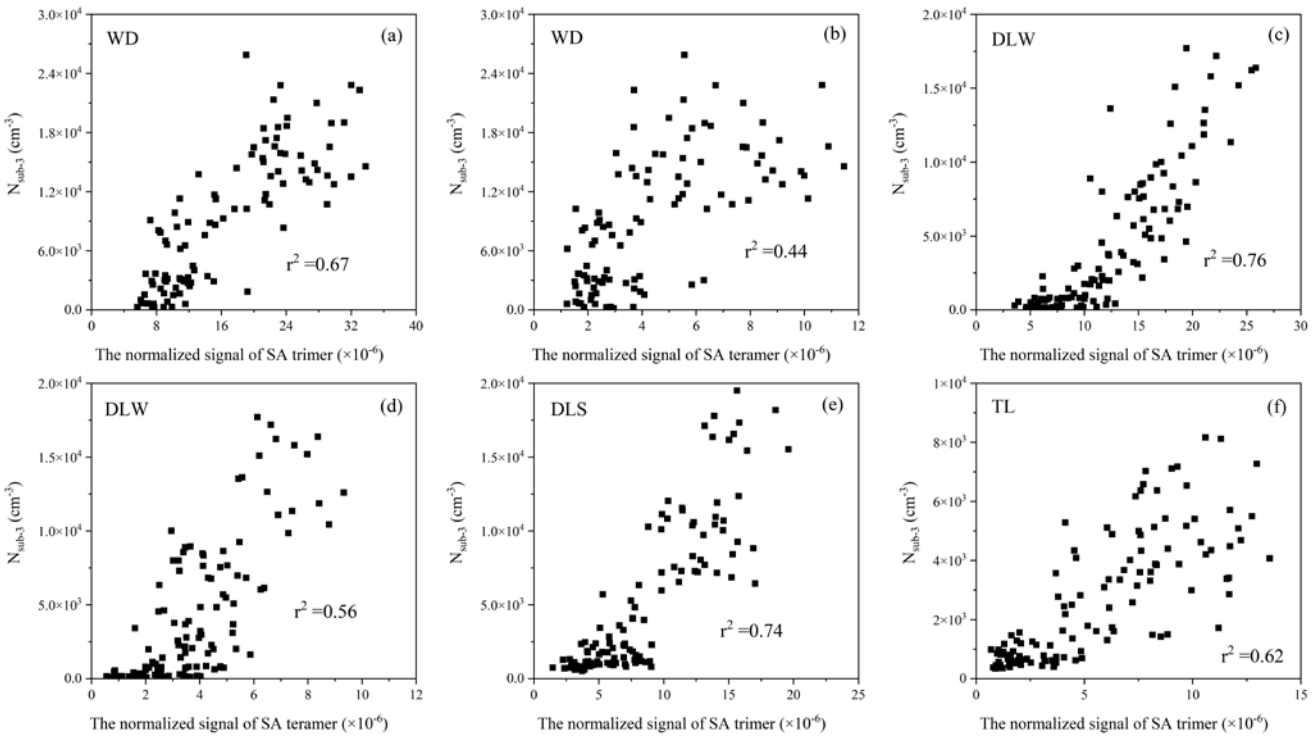

Figure S3: The correlation between the normalized signal of SA clusters and the number concentration of sub-3 nm particles: (a)-(b) January 20th, 2019 at WD; (c)-(d) January 27th, 2023 at DL in winter; (e) May 2nd, 2023 at DL in spring; (f) August 7th, 2023 at TL. Only data recorded between 06:00 and 18:00 were included in the analysis."

We have also provided a description of this figure (lines 190-193):

"Besides, the normalized signal of SA trimer and tetramer show good correlations (r^2 = 0.44-0.76) with the number concentration of sub-3 nm particles (Figure S3). Such correlations between clusters and newly formed particles were reported in other atmospheric observations (Bianchi et al., 2016; Yan et al., 2021), indicating that these clusters

typically signified the molecular clustering processes in NPF events."

For S-O ions, they are clusters containing only sulfur/oxygen atoms beyond sulfuric acid. we have removed them, as they are not directly relevant to NPF, and retained SA and SA-DMA clusters in the figure. The area of symbol is proportional to cluster signals, and we have also annotated the logarithm of normalized signals in this figure. The revised Figure 2 is shown as (lines 197-204):

"

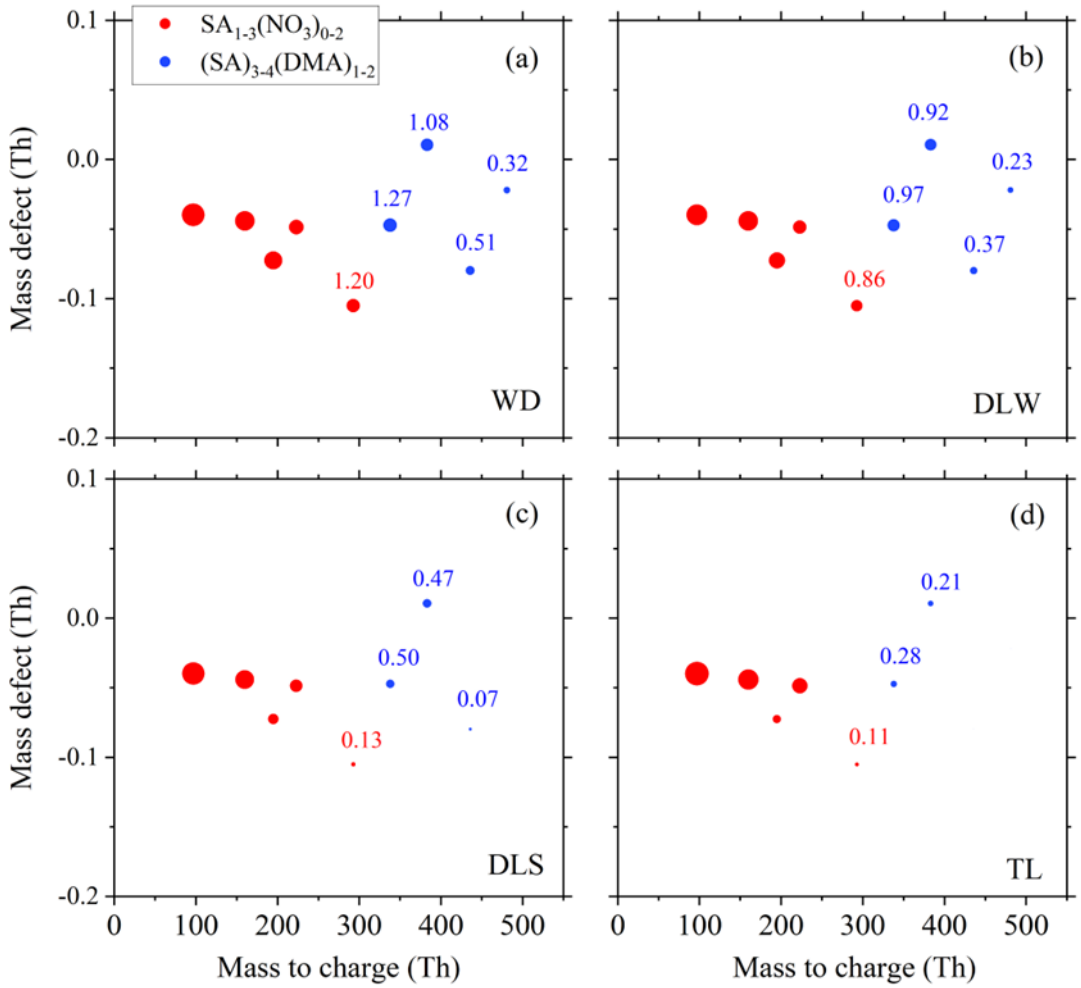

Figure 2: The mass defect of SA molecules and its clusters during four NPF events on (a) January 20th, 2019 at WD (temperature = 275 K; [SA_1] = 1.4×10^7 cm^{-3}; CS =0.055 s^{-1}; [DMA] = 3.4 pptv); (b) January 27th, 2023 at DLW (temperature = 277 K; [SA_1] = 7.3×10^6 cm^{-3}; CS =0.012 s^{-1}; [DMA] = 1.5 pptv); (c) May 2nd, 2023 at DLS (temperature = 295 K; [SA_1] = 9.5×10^6 cm^{-3}; CS =0.014 s^{-1}; [DMA] = 2.9 pptv); (d) August 7th, 2023 at TL (temperature = 304 K; [SA_1] = 2.6×10^7 cm^{-3}; CS =0.023 s^{-1}; [DMA] = 1.8 pptv). Other species detected by CI-LToF-MS were not shown, because they are not directly related to atmospheric nucleation. The area of symbol is proportional to the logarithm of normalized signal. (multiplied by a factor of 1×10^6 before taking the logarithm). The logarithm of values is annotated for larger clusters."

4. Sentence on line 245 talks about dependence on DMA and mentions Figure S4 but that figure shows no (or even inverse!) difference in [DMA] between events and non-events.

Response:

Regarding the difference in [DMA] between events and non-events, we think the referee referred to Figure S4 (formerly Figure S2). In fact, the dependence of NPF on [DMA] are not contradictory to the difference in [DMA] between events and non-event. As discussed above (in the response to Comment #2.2), there was a good correlation between [DMA] and CS during NPF periods, and the suppression of NPF by high CS likely masked the enhancing effect of DMA. We have a new figure (Figure S6), which shows a positive correlation between [DMA] and CS (lines 186-188 in the Supplement):

"

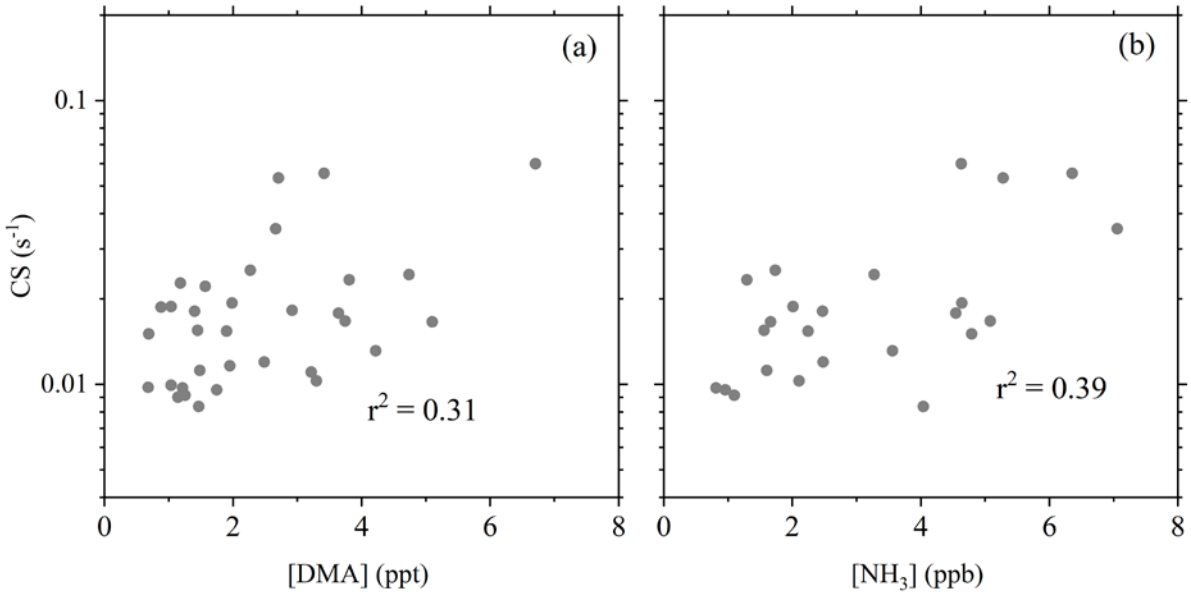

Figure S6: Correlations between the concentrations of basic precursors and CS during NPF periods: (a) [DMA] at WD, DL and TL; (b) [NH_3] at WD and DL. The NPF period is defined as the period with the maximum value of $J_{1.7}$ in each NPF event."

The dependence of NPF on [DMA] becomes discernible when other factors are accounted for, as evidenced in Figure S5 (formerly Figure S4).

5. There are many sentences with strange wording choices e.g. in paragraphs such as lines 59-70, lines 138-150 (also, this scaling procedure leaves the reader a bit uneasy).

Response:

The manuscript has been revised for better expression, logical clarity, and grammatical precision. All major co-authors contributed to this revision.

6. Lines 304-315 in the conclusions are accurate and reflect the indirect nature of the evidence for SA-DMA nucleation but what does this paper add to what is already there? It seems the present day tools are inadequate to directly resolve the question. What should be improved? Looking for some more strongly worded conclusions.

Response:

Thanks for referee's comments. We have improved the Conclusion section to better highlight our key findings.

Using multi-site data from eastern China, our primary contribution lies in demonstrating that the SA-DMA mechanism remains applicable across broader spatial and temperature ranges. Furthermore, we provide atmospheric evidence supporting the role of OOMs in the initial growth of new particles. The revised version is (lines 348-368):

"We studied the mechanism and influencing factors of NPF on a large spatial scale by using observational data at multiple sites in the eastern region of China, including the concentrations of key chemical species, temperature and PSD.

By Comparing with previous studies that investigated atmospheric nucleation mechanism in individual sites, we have showed the applicability of a similar mechanism over a large geographic region. Based on the correlation between [SA_1] and particle formation rate, the identification of key clusters, and comparisons between simulations and measurements, we concluded that nucleating processes, mainly exemplified by SA_2 and $J_{1.4}$, could be largely attributed to SA-DMA collision, although other precursors, such as NH_3 and OOMs, might also participated. However, SA and its clusters were insufficient, at least at DSL and BJ, to explain the initial growth of nucleated particles, while OOMs make great contribution to this process, thereby affecting $J_{1.7}$, which was derived from model-observation comparisons. While previous understanding of the contribution of OOMs to initial particle growth was reflected by chamber studies, our study provides supporting evidence from atmospheric observations. Given the considerable spatial separation among five sites, we infer that within this extensive urban agglomeration, SA and DMA are capable of describing atmospheric nucleation up to 1.4 nm, whereas OOMs are potentially involved in in subsequent growth. This may also apply to other populated and polluted regions, where the NPF mechanism warrants investigation.

As for influencing factors, the occurrence of NPF was governed by [SA_1] and CS, whereas the frequency and intensity of NPF were mainly determined by temperature, which generally exhibited a negative correlation with NPF over a wide temperature range. Compared with previous studies investigating NPF dependence on temperature from a temporal perspective, dependences of cluster concentrations and particle formation rates on temperature were illustrated quantitatively through scaling at five sites, suggesting the differences in nucleation intensity across this region could be explained by variations in temperature under the comparable mechanism. We expect that this finding holds for similar polluted atmospheric environments on large spatial scales worldwide, particularly where significant temperature gradients exist."

This manuscript is based on likely state-of-the-art analytical approaches and the best currently available dataset for nucleation in polluted atmospheres. By applying a scaling approach to account for multiple atmospheric factors, we have provided multiple evidence supporting SA-DMA nucleation. However, as the referee noted, these pieces of evidence remained indirect. We think that future advances in the direct measurement of nucleating clusters are essential to reach more definitive mechanistic conclusions, which may lead to the unveiling of the contribution from other nucleation precursors. Our discussion of future research directions has been revised as (lines 369-374):

"The case for SA-DMA nucleation presented in this study remains indirect, as it relies on precursors, limited clusters and modeling rather than comprehensive identification of nucleation process. To better resolve atmospheric nucleation mechanisms, future studies are warranted to advance beyond current methods of cluster detection, for

instance, by enabling direct measurements of all basic molecules within SA clusters. In the complex urban atmosphere, the potential involvement of other pollutants will require novel direct measurements and analytical techniques, along with more comprehensive modeling, to clarify the roles of additional precursors."

We appreciate the referee for these constructive comments, which have significantly improved the quality and clarity of our manuscript.

Anonymous referee #2:

*1. The paper by Jin et al. examines the role of different molecules in NPF at various sites in China. While the authors provide valuable measurements of ammonia and amine concentrations—data that are otherwise scarce—the presentation of their work and the evidence supporting their conclusions are weak. In its current form, the paper does not add substantial new findings to the literature, although the analysis of temperature effects on DMA concentrations and clustering is novel, interesting, and important. However, the manuscript often reads as if it is written primarily for scientists within China, as it includes almost no comparisons with sites outside of China (with one unclear exception). The same issue arises with self-citations, which dominate the references to existing literature. In several cases, conclusions are drawn from figures where the evidence is either unclear or missing. The title also does not accurately reflect the paper's findings. In addition, the manuscript is difficult to follow, with several contradictory statements. I recommend that the authors substantially revise the manuscript: clarify the contradictions, sharpen the focus on their novel findings, and compare their results to observations from outside China. It is also essential to strengthen the methods section so that the results can be reproduced in other locations. Below, *some* specific comments are listed.*

Response:

We sincerely thank the referee for recognizing the challenges associated with our data scarcity and acknowledging our work. Based on these comments, we have conducted a major revision of the presentation, new findings, and methods of our work. Below, we reorganize the referee's comments above and respond to them point by point.

1.1. The presentation of their work and the evidence supporting their conclusions are weak. In its current form, the paper does not add substantial new findings to the literature. In addition, the manuscript is difficult to follow, with several contradictory statements.

Response:

Thank the referee for pointing these out. We think that these issues mainly stemmed from the imprecise wording and inadequate explanation of evidence, and we have therefore revised the manuscript accordingly.

Regarding the presentation of our work and the evidence supporting our conclusions, we have made the following major revisions:

(1) In the Introduction, we have expanded the emphasis on the mechanisms and influencing factors of NPF in a broad region, while streamlining the focus on eastern China to present a global perspective.

(2) In the Results and Discussion, we have also incorporated field observations from various global sites to enrich the discussion. We have also provided and clarified evidence for SA-DMA nucleation.

(3) In the Conclusions, the section has been thoroughly revised to better highlight the novel contributions of our work. We have also outlined prospects for future research into NPF mechanisms.

Regarding new findings, this study investigates the consistency of NPF mechanisms across an extensive region and demonstrates that observed differences in NPF within the region are attributed to temperature variations, rather than to changes of the nucleation mechanism itself. The revised Conclusion Section includes comparisons with prior studies, which highlights novel findings, summarized as follows:

(1) SA and DMA can explain atmospheric nucleation across large spatial scales, whereas previous research on SA-DMA nucleation was confined to individual sites.

(2) OOMs are necessary to explain the growth rates of newly nucleated particles. This provides crucial atmospheric observational evidence, whereas previous understanding of the contribution of OOMs to initial particle growth was derived from chamber studies.

(3) Temperature is the governing factor for determining NPF characteristics across various sites, whereas previous atmospheric observations mainly focused on explaining its effects over temporal scales.

Regarding contradictory statements, in addition to addressing the contradictions highlighted in Comment #9, Comment #10 and Comment #16, further revisions include:

(1) An explanation for the discrepancies between measured and simulated values in Figure S7 (lines 127-132 in the Supplement): "There is an acceptable consistency between $J_{1.4,meas}$ and $J_{1.4,sim}$ when observational and model uncertainties are considered (Figure S7). Deviations reported in previous laboratory or field studies were typically within one order of magnitude (Kürten et al., 2018; Cai et al., 2021; Xiao et al., 2021), smaller than those in our study. This discrepancy likely arises from differences in uncertainty ranges. For a single observation, the measurement uncertainty is relatively low (Freshour et al., 2014), and the resulting simulation uncertainty can be controlled within a narrow range. However, our measurement spanned several years and involved multiple sites, may amplifying the overall uncertainty in $J_{1.4}$."

(2) A discussion of cases where NPF exhibits weak temperature dependence (lines 299-309): "When temperature differences are substantial, its inhibitory effect on NPF becomes evident in complex ambient environments. For example, Baalbaki et al. (2021) reported the particle formation rate in warmer months was actually lower than that in cooler months with comparable [SA_1] levels in the Eastern Mediterranean. Inter-site comparisons in India showed a negative correlation between particle formation rate and temperature, although precursor concentrations were not measured (Kanawade et al., 2022). However, under conditions of limited temperature variability, the effect of temperature is less directly observable, and higher temperatures may even appear to favor NPF (Größ et al., 2018; Brean et al., 2020; Yan et al., 2021; Victor et al., 2024). This may stem from other temperature-related factors. For example, in atmospheric observations, temperature is often correlated positively with solar radiation, which directly promotes the formation of nucleation precursors (Kürten et al., 2016; Brean et al., 2020). Additionally, temperature is typically negatively correlated with relative humidity, and high humidity suppresses NPF via hygroscopic growth of pre-existing particles

(Määttänen et al., 2018).”

(3) The content (formerly lines 261-265) that contradicted point #2 above, as well as the corresponding figure (formerly lines 147-149 in the Supplement) have been deleted.

1.2. Almost no comparisons with sites outside of China (with one unclear exception). The same issue arises with self-citations.

Response:

Thanks, we have compared the findings from sites outside China with the results of this study. References on these studies have been added, and unnecessary self-citations have been removed.

1.3. In several cases, conclusions are drawn from figures where the evidence is either unclear or missing.

Response:

We thank the referee for pointing this out. All figure-derived conclusions have been checked. We have revised:

(1) The conclusion drawn from Figure 1 (lines 172-173) “Therefore, the observed nucleation is likely driven by SA and enhanced by strong stabilizing precursors such as DMA. Other nucleation mechanisms are unlikely to dominate under these conditions.”

(2) The conclusion drawn from Figure 4 and Figure S7 (lines 253-254) “The consistency between measured and simulated parameters ([SA_2] and $J_{1.4}$) supports the significance of SA-DMA collision in NPF.”

(3) The conclusion drawn from Figure 7 (lines 328-329) “The comparison between $J_{1.7,meas}$ and simulated $J_{1.7}$ ($J_{1.7,sim}$) provides support for the contribution of OOMs to the growth of nucleated particles (Figure 7b).”

1.4. The title also does not accurately reflect the paper’s findings.

Response:

The findings of this study are given in our response to Comment #1.2. The original title, “Atmospheric new particle formation in the eastern region of China: a mechanistic investigation at multiple sites,” reflected only two of these contributions and overlooked the analysis of influencing factors. To address this omission, the title has been revised to “Atmospheric new particle formation in the eastern region of China: an investigation on mechanism and influencing factors at multiple sites”.

1.5. It is also essential to strengthen the methods section so that the results can be reproduced in other locations.

Response:

To strengthen the methods section, measurement details of SA and OOMs have been added (lines 8-19 in the Supplement):

“For the measurement of sulfuric acid (SA) and oxygenated organic molecules (OOMs), a sample flow (8.8 L min^{-1}) was positioned centrally along the axis of the nitrate chemical ionization source, encased by a sheath gas flow (20 L min^{-1}) consisting of high-purity air containing trace gaseous nitric acid to minimize wall loss of analyte molecules. Under the influence of an electric field, nitrate ions were driven toward the center of the sample flow and charged sample molecules. After chemical ionization, approximately 0.8 L min^{-1} of the ionized sample flow was directed through a 0.3 mm pinhole into three successively evacuated chambers under differential pressure and guided electric fields. [SA] was determined by (Lu et al., 2019):

$$[SA] = C_{SA} \cdot \frac{(HNO_3)_{0-2}SA^- + SA_2^-}{(HNO_3)_{0-2}NO_3^-} \quad (S1)$$

where C_{SA} is the calibration coefficient for SA; $(HNO_3)_{0\text{-}2}SA^-$, SA_2^- and $(HNO_3)_{0\text{-}2}NO_3^-$ represent the signals (cps). The fractional part on the right side represents the normalized signal of SA. [OOMs] was determined by (Kirkby et al., 2016):

$$[OOM_X] = \frac{C_{SA}}{T_X} \cdot \frac{OOM_X \cdot NO_3^-}{(HNO_3)_{0-2}NO_3^-} \quad (S2)$$

where T_X is the mass-to-charge-dependent transmission efficiency relative to SA; $OOM_X \cdot NO_3^-$ represent the signals (cps).”

For the detailed information of measuring DMA and NH_3, please refer to our response to Comments #5 below.

Specifically:

2. Article needs general grammar check e.g.

Line 25: ‘causes’ to be changed to ‘cause’ or ‘result in’

Line 67: sentence starting with ‘In particular,’ needs modification

Please make sure to use a consistent verb tense throughout the manuscript.

Response:

Thanks, we have checked the grammar and verb tense throughout the manuscript.

3. A map of the sites, including distances and major emission source locations (if possible), could benefit the readers. Please add it to the supplementary material.

Response:

A map of the sites has been provided (lines 155-158 in the Supplement):

“

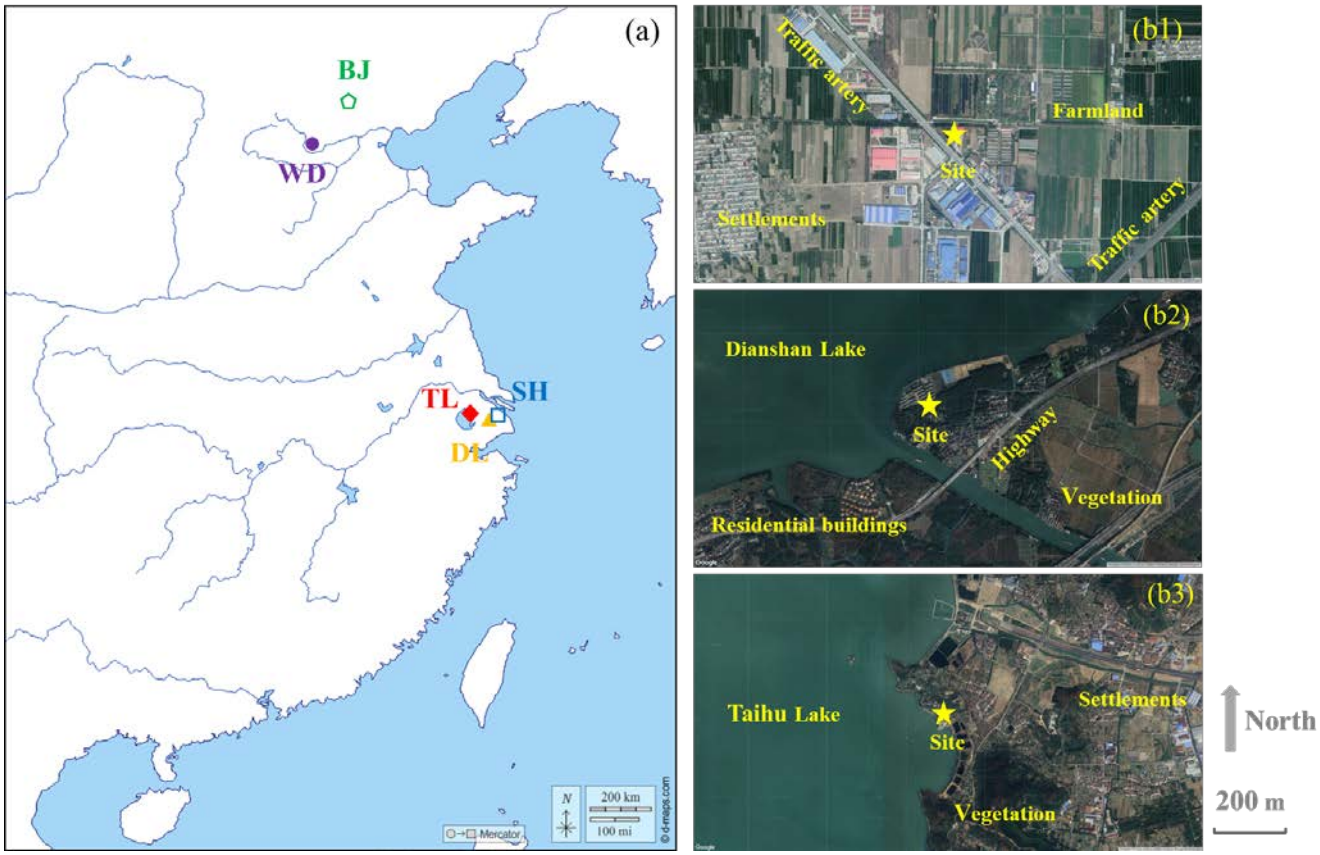

Figure S1: The location of the measurement sites: (a) The overall distribution across eastern China; (b1) WD; (b2) DL; (b3) TL. The location of Beijing site (BJ) and Shanghai site (SH) was introduced in the corresponding studies (Yao et al., 2018; Cai et al., 2021).”

4. Line 85, is it the same mass spectrometer moved between the locations? How are the authors sure that the moving of the spectrometer does not affect the calibration factor and m/z calibration.

Response:

Yes, we used the same mass spectrometer at these three sites. Instrument calibrations were conducted before each measurement to maintain the accuracy of the data. We have revised our manuscript (lines 90-94) as:

“A calibration coefficient derived from SA and a mass-to-charge-dependent transmission efficiency of the instrument were used to obtain [OOMs], assuming that they share the same kinetically controlled collision rate with reagent ions as that of SA. The transmission efficiency was calibrated using a system coupling a high-resolution differential mobility analyzer (HR-DMA) with the mass spectrometer (Heinritzi et al., 2016). The calibrations of [SA] and transmission efficiency were performed before each campaign.”

5. Line 89: I suggest to the authors to add details about the measurements of amines using the ToFs since these are new types of measurements, and it would be good to make the method reproducible. Line 94: More info on the IC measurements is needed since the text is not available in English.

Response:

Thanks, the measurement methods for two alkaline gases have been added (lines 20-46 in the Supplement):

"For the measurement of dimethylamine (DMA) at Wangdu (WD) and Dianshan Lake (DL), the Vocus proton-transfer-reaction time-of-flight mass spectrometer generated protonated water ions through specific voltage (450 V) and current (1.4-1.5 A) settings. The ion source introduced ultra-pure water vapor (20-30 sccm) into focusing ion-molecular reactor (FIMR), where it underwent proton transfer reactions with DMA. Efficient ion-molecule reactions were ensured by a precise control of temperature (100 °C) and pressure (2 mbar). The electric field strength in the FIMR was set to 170 Td, effectively focusing and accelerating the generated ions. The radio frequency amplitude and frequency in big segmented quadrupole were set at 240 V and 2.2×10^6 Hz, respectively. At WD, DMA standard gas was generated by a self-made permeating tube. The permeation rate of the tube was determined by an acid-base calibration. At DL, six species (methyl ethyl ketone, benzene, toluene, m-xylene, 1,3,5-trimethylbezene and α-pinene) were calibrated to obtain the sensitivity for DMA, as the fitting ratio of proton-transfer reaction rate constants to sensitivity of these gases is very close to that of DMA (Wang et al., 2020). The ambient mixing ratio of [DMA] at WD and DL was calculated by (Krechmer et al., 2018):

$$[\mathrm{DMA}] = \frac{I}{\mathrm{S_{DMA}}} \quad (S3)$$

where I is the signal of $C_2H_8N^+$ (cps); S_{DMA} is the sensitivity for DMA.

For the measurement of DMA at Taihu Lake (TL), a pure air flow (1 L min^{-1}) was directed through an ethanol-filled bubbler and subsequently entered a radioactive source to generate protonated ethanol reagent ions. A sample flow (1.35 L min^{-1}) was introduced into the ion-molecule reaction (IMR) chamber, where it mixed with the reagent ion. The pressures in the IMR chamber and the small-segmented quadrupole were maintained at approximately 100 mbar and 2.8 mbar, respectively, to enhance instrument sensitivity. To minimize wall losses, a high sampling flow rate (15 L min^{-1}) was employed, and the sampling line was heated to 50 °C. The calibration method was similar to that at WD. The ambient mixing ratio of [DMA] at TL was calculated by (Yao et al., 2016):

$$[\mathrm{DMA}] = \mathrm{C_{DMA}} \cdot \frac{(\mathrm{DMA})(\mathrm{C_2H_5OH})_{0-1}\mathrm{H^+}}{(\mathrm{C_2H_5OH})_{1-3}\mathrm{H^+}} \quad (S4)$$

where C_{DMA} is a calibration coefficient for DMA. $(DMA)(C_2H_5OH)_{0\text{-}1}H^+$ and $(C_2H_5OH)_{1\text{-}3}H^+$ represent the signals (cps).

For the measurement of NH_3, the sampling process involved drawing ambient air at a flow rate of 0.5 L min^{-1} for 60 minutes through a 0.45 μm filter and into a 10 mL SA absorption solution (0.01 mol L^{-1}) held in a porous glass absorber, with a typical sample volume of 30 L. After sampling, the absorption solution was transferred to a 10 mL tube, diluted with water, and then filtered through a 0.22 μm membrane. The analysis was performed using ion chromatography with a cation exchange column and a conductivity detector. The quantification of [NH_3] was achieved using an external standard method."

6. Line 124: please list the different errors

Response:

A corresponding table has been added (line 207 in the Supplement):

“**Table S1: The range of uncertainty associated with parameters in models.**

Input	[SA]	[DMA]	[OOMs]	CS	Particle formation rate
Uncertainty	+100%/-50%	+150%/-60%	+200%/-66% (logC^*±1)	±10%	+100%/-50%

”

7. Table 2: Data source is misspelled

Response:

The spelling errors have been corrected.

8. How does the frequency of NPF events differ between the sites?

Response:

The frequency of NPF events shows a strong temperature dependence between the sites. A bar chart representing NPF frequency has been added in Figure 3 (panel d) and a discussion of NPF frequency has been provided (lines 228-232):

“Although the occurrence of NPF is not strongly dependent on ambient temperature in individual campaigns (Figure 3c), the intercomparison across campaigns indicates a general negative correlation between NPF frequency and temperature, reflecting seasonal characteristics (Figure 3d). This trend is consistent with long-term measurements in Chacaltaya (Rose et al., 2015), Beijing (Deng et al., 2020) and Gwangju (Lee et al., 2024), where NPF was most frequent in winter and least frequent in summer.”

“

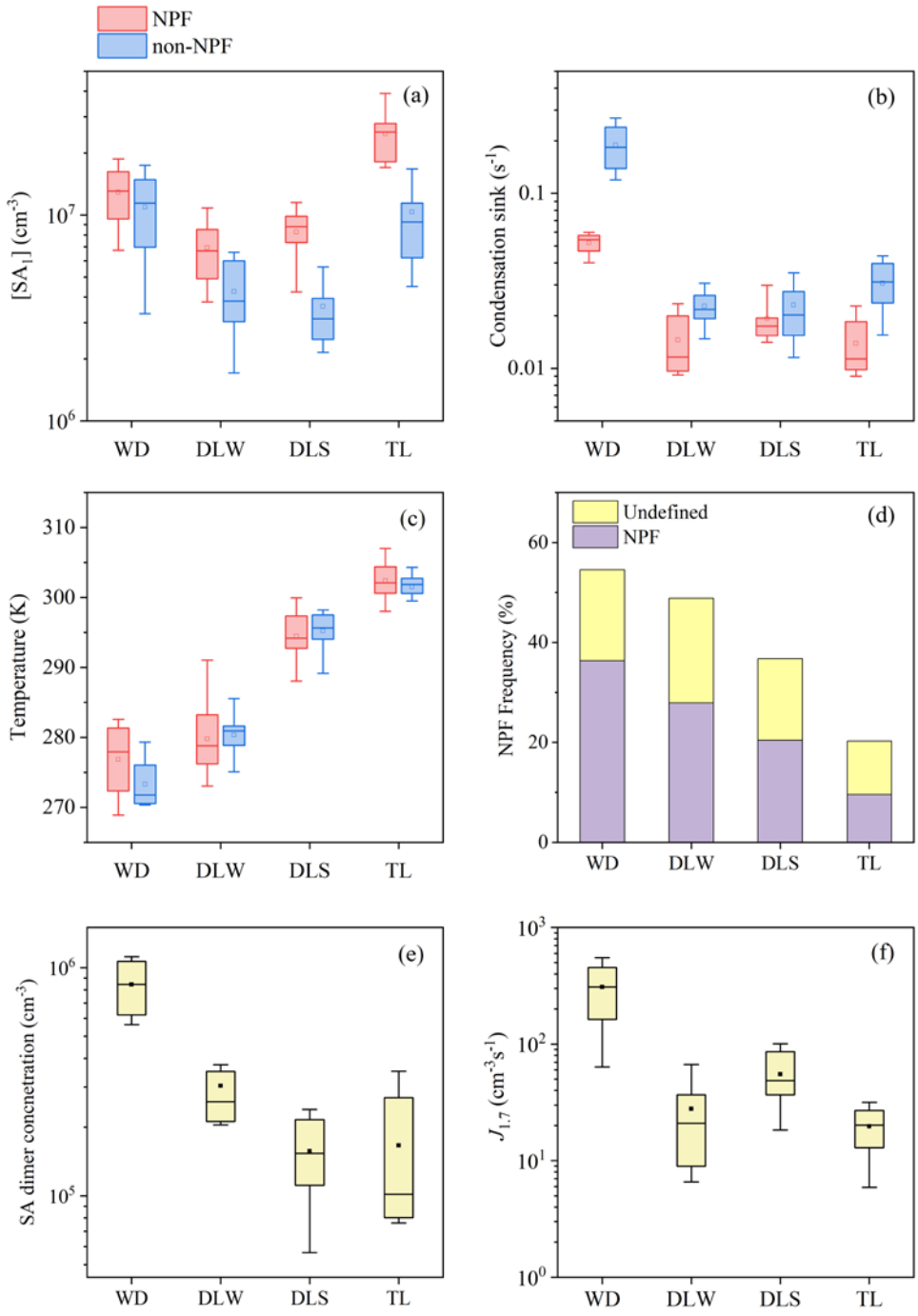

Figure 3: Parameters related to NPF. (a) [SA_1], (b) CS, (c) temperature during NPF periods and non-NPF periods, (d) NPF frequency in each campaign, and (e) [SA_2] and (f) $J_{1.7,meas}$ during NPF periods. The NPF period is defined as the period with the maximum value of $J_{1.7}$ in each NPF event, and the non-NPF period is defined as the median range of all NPF periods (9:00-11:00) in non-NPF days. In order to eliminate the influence of precipitation, only sunny and cloudy days are selected for non-NPF. The transverse lines and square markers inside the boxes indicate mean values and median values, respectively. The bottom and top edges of the box indicate the 25th and 75th percentiles, respectively. The bottom and top edges of the whisker lines outside of the boxes indicate the 10th and 90th percentiles, respectively.”

9. Line 164-165: The statements need clarification; how can the authors be sure that the mechanism is DMA-SA driven although they mention that it is unclear whether OOMs and NH_3 participate in the nucleation.

Response:

Sorry for the inaccurate statements. We suggest that SA and DMA play key roles in nucleation, while not ruling out the contribution of other species (OOMs and NH_3) to cluster growth. We have revised the original statement "the dominate NPF mechanism in our campaigns was identified as SA-DMA" into "the observed nucleation is likely driven by SA and enhanced by strong stabilizing precursors such as DMA. Other nucleation mechanisms are unlikely to dominate under these conditions" (lines 172-173) and the contribution of OOMs was in Sect. 3.4.

10. Figure S2 does not support this conclusion, how do the authors explain the higher DMA and NH_3 on non-NPF days?

Response:

This is a misunderstanding due to the lack of explanation of this apparent contradiction. Although DMA is recognized as one of the key nucleation precursors, its role cannot be reflected simply by comparing [DMA] between NPF days and non-NPF days. The reason lies in the correlation between [DMA] and other influencing factors, especially the positive correlation between [DMA] and CS, where the inhibitory effect of CS on NPF outweighs the promoting effect of DMA. It is likely for the same reason, [NH_3] was high on high-CS days, resulting the negative correlation between [NH_3] and the occurrence of NPF. We have revised the manuscript as (lines 218-227):

"Increasing [DMA] considerably enhances SA_2 formation under fixed temperature ranges (Figure S5), and this effect diminishes as [DMA] approaches nucleation saturation (Almeida et al., 2013). This phenomenon supports the contribution of DMA to atmospheric nucleation and is consistent with the results from a flow reactor (Jen et al., 2014). In CLOUD studies, the contributions of OOMs and NH_3 to cluster formation were almost negligible in the presence of DMA (Kürten et al., 2018; Xiao et al., 2021). Given that our field measurements exhibited similar [NH_3] and relatively low [OOMs] compared to those in CLOUD experiments (Table 2), it follows that their effects are also minor under our ambient conditions. Despite the potential participation of other precursors, the observation-simulation agreement suggests DMA is a major base that stabilizes SA clusters (Figure S5). However, [DMA] and [NH_3] were not markedly elevated during NPF periods (Figure S4). The likely reason is that the suppression of NPF by high CS masked the enhancing effect of DMA and NH_3, as they were positively correlated with CS (r^2=0.31 and 0.39, respectively, Figure S6)."

11. Figure 2: are the shown molecules the only ones observed in the CIMS, or did the authors use a specific filter for the other peaks?

Response:

The figure shows only the clusters containing the SA molecule. Other detected ions are not shown, as they are not directly related to atmospheric nucleation. We have revised our manuscript (lines 201-202) as: "Other species detected by CI-LToF-MS were not shown, because they are not directly related to atmospheric nucleation."

12. Line 200: why do the authors think that CS affects NPF at the measured locations but not in the PoValley?

Response:

Our original statement might be confusing. While the CS affects NPF at both the Chinese sites in this study and the Po Valley site, it was likely a governing factor on the occurrence of NPF at the measured locations, but not in the Po Valley. We have revised our manuscript (lines 211-214) as:

"CS during NPF periods was generally lower than that during non-NPF periods in all campaigns, except DLS, further confirming that high preexisting aerosols suppressed the occurrence of NPF (Figure 3b). However, NPF events were not significantly dependent on the CS in Po Valley, indicating other influencing factors govern the occurrence of NPF in less polluted atmospheres (Cai et al., 2024)."

13. Line 200: this is the first (and only) mention of a location outside of China. The authors are encouraged to widen the comparison to polluted locations outside of China, and to refer to studies of other groups working on the topic.

Response:

Thanks. We have added the discussion with observational data from multiple international studies, including work by other research groups, covering both polluted and clean environments.

14. Line 202: do these vapors affect the growth, or intensity of nucleation? Please clarify.

Response:

We have provided a description of how these vapors affect NPF (lines 216-218):

"Besides SA, other potential precursors related to NPF in polluted regions including DMA, NH_3 and OOMs (Figure S4). These species are known to enhance SA-driven nucleation (Kirkby et al., 2011; Almeida et al., 2013; Riccobono et al., 2014) and promote particle growth (Tröstl et al., 2016)."

15. Line 210: please clarify the role of temperature, did it influence the intensity, occurrence, or concentration of vapors?

Response:

The influence of temperature on the occurrence and intensity of NPF has been revised (lines 228-230 and lines 232-237, respectively):

"Although the occurrence of NPF is not strongly dependent on ambient temperature in individual campaigns (Figure 3c), the intercomparison across campaigns indicates a general negative correlation between NPF frequency and temperature, reflecting seasonal characteristics (Figure 3d)." and

"The intercomparison of campaigns further shows that temperature played a key role in determining the intensity of NPF indicated by [SA_2] and $J_{1.7,meas}$ (Figure 3e-f). Temperature even exerted more dominant influence than [SA1] and CS. For example. despite the highest [SA_1] at TL, its elevated temperatures (298-306 K) resulted in lower median values of [SA_2] and $J_{1.7,meas}$ than those in all campaigns. Conversely, WD had the lowest temperature (281-

268 K) and relatively high [SA_1] (7×10^6-1.8×10^7 cm^{-3}), the intensity of NPF at WD was the highest, even under strong coagulation scavenging (median CS = ~0.05 s^{-1}). A more detailed discussion regarding the influence of temperature on NPF is provided in Sect. 3.3."

The effect of temperature on the concentration of vapors may be somewhat beyond the scope of that paragraph, which has been moved to lines 304-308 in the Main Text and lines 149-153 in the Supplement for further discussion: "However, under conditions of limited temperature variability, the effect of temperature is less directly observable, and higher temperatures may even appear to favor NPF (Größ et al., 2018; Brean et al., 2020; Yan et al., 2021; Victor et al., 2024). This may stem from other temperature-related factors. For example, in atmospheric observations, temperature is often correlated positively with solar radiation, which directly promotes the formation of nucleation precursors (Kürten et al., 2016; Brean et al., 2020)." and

"The second is the effect on the volatility of individual OOMs species (Epstein et al., 2010), corrected using Equation S10. The third is the effect on organic oxidation rates, which alter the number of effective oxygen of OOMs to change their volatilities. This path involves the whole system of organic oxidation in the atmosphere and is beyond the scope of this study. The third effect has been identified to be weaker than the second one in a chamber investigation (Caudillo et al., 2021), and can be relatively neglected to some extent."

16. Line 292: contradicts lines 164-165, please clarify.

Response:

Based on the available data and analysis, our conclusions do not rule out the involvement of OOMs in NPF, and we emphasize that SA and DMA can explain atmospheric nucleation. As for OOMs, we subsequently demonstrate their theoretical capacity to influence the initial growth of particles in later sections.

We have revised 172-173 lines (formerly lines 164-165) as: "Therefore, the observed nucleation is likely driven by SA and enhanced by strong stabilizing precursors such as DMA. Other nucleation mechanisms are unlikely to dominate under these conditions."

We have also revised lines 334-337 (formerly line 292) as: "In fact, some ultralow volatile organic compounds within OOMs may be involved in nucleating (Simon et al., 2020). Nevertheless, even without considering their contribution in this stage, the condensation of OOMs exerts a considerable influence on initial growth, as reflected in $J_{1.7}$. Moreover, $J_{1.7}$ exhibits strong temperature dependence, similar to $J_{1.4}$ (Figure S9)."

17. Line 314: Do the authors mean that the production of SA depends on SO_2, and not the other way around?

Response:

Yes, the production of SA depends on SO_2. Following the removal of the discussion on future nucleation mechanisms, this sentence (formerly line 314) is no longer included in the manuscript.

18. Figure S7: What information do the authors infer from the volatility distribution? There is too little text explaining this figure in the manuscript. If there is no use of this figure (or data) elsewhere, the authors are encouraged to remove this part from the manuscript.

Response:

We agree and have deleted this figure.

We express our sincere thanks to the referee for the valuable comments, which have greatly improved this manuscript.