A point-to-point response to referee #2

We are very grateful for your constructive comments, and have carefully revised our manuscript accordingly. A point-by-point responses to the comments is given below. Your 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.

*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 your 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 your 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 you 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 you 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):

“

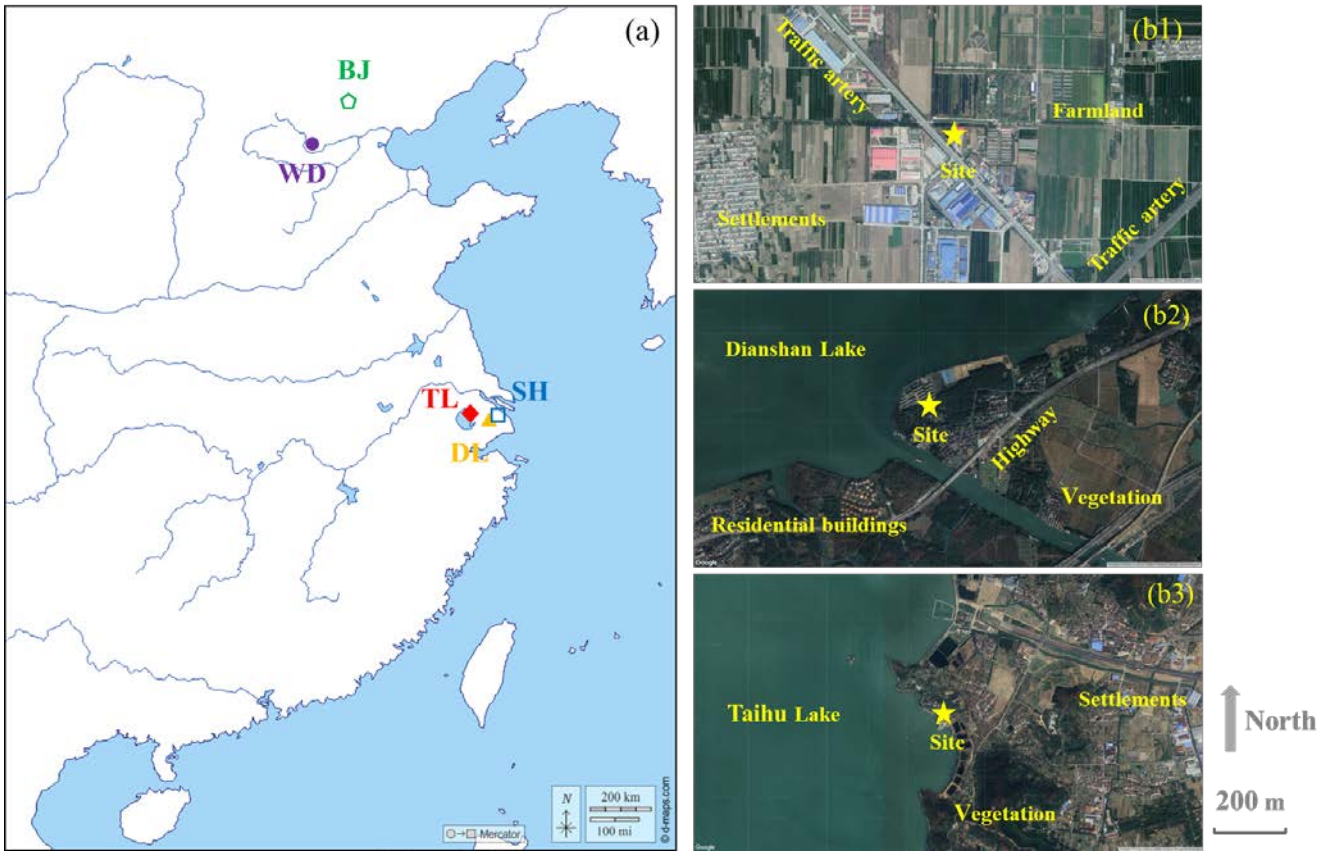

Figure S1: The location of the measurement sites: (a) The overall distribution across eastern China; (b1) WD; (b2) DL; (b3) TL. The location of Beijing site (BJ) and Shanghai site (SH) was introduced in the corresponding studies (Yao et al., 2018; Cai et al., 2021).”

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."

“

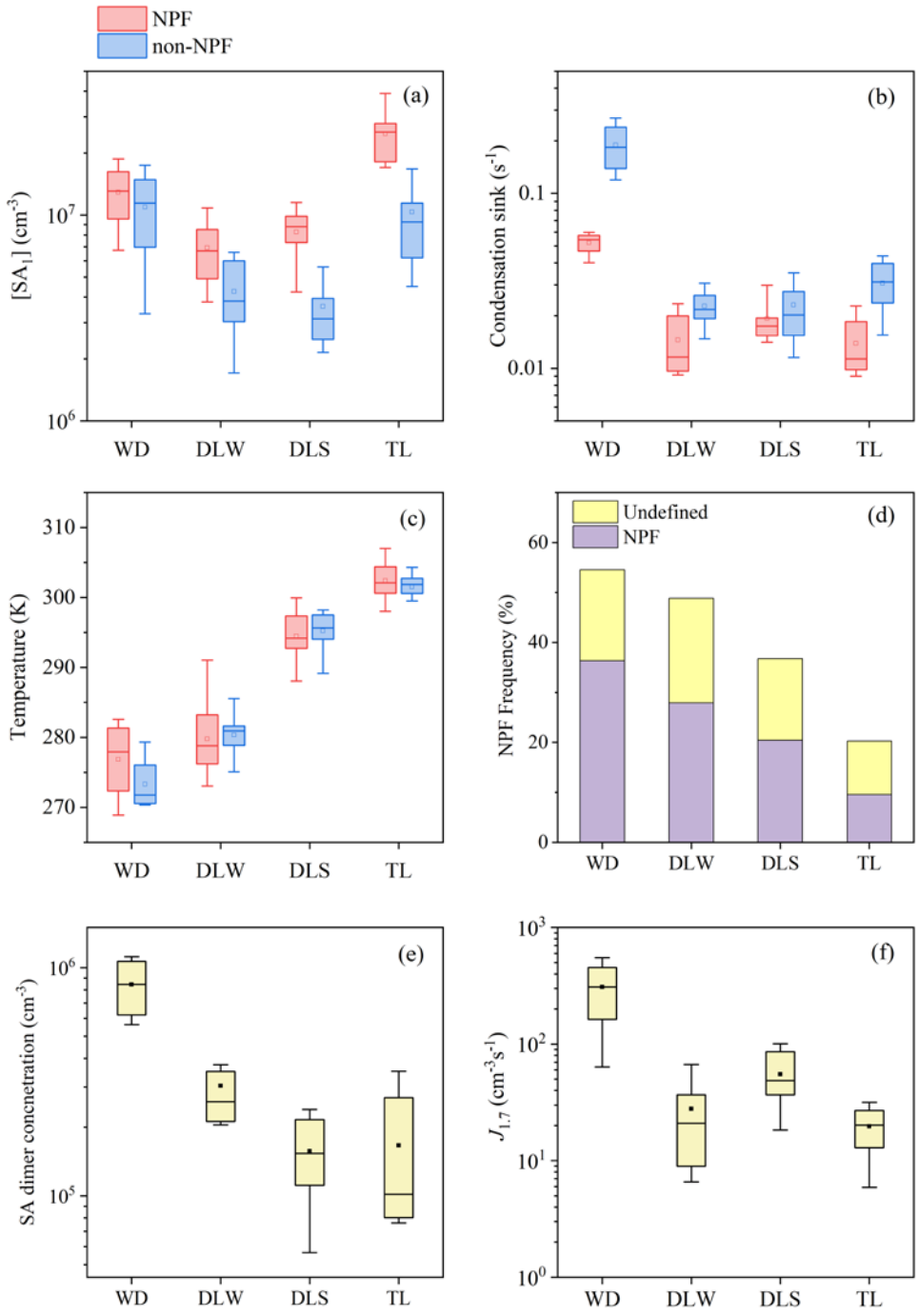

Figure 3: Parameters related to NPF. (a) [SA_1], (b) CS, (c) temperature during NPF periods and non-NPF periods, (d) NPF frequency in each campaign, and (e) [SA_2] and (f) $J_{1.7,meas}$ during NPF periods. The NPF period is defined as the period with the maximum value of $J_{1.7}$ in each NPF event, 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 you for the valuable comments, which have greatly improved this manuscript.