

A point-by-point reply to both referees' comments

We thank the constructive comments from the referees. A point-by-point reply to the comments, which are reproduced in *italics*, is provided below. Our responses are given in normal fonts. Line numbers in the responses correspond to those in the revised manuscript. Revised text is shown in blue.

Referee #1:

The authors have done a laudable job in revising the manuscript. The final paragraph is welcome indeed. Three important points:

Response:

We sincerely appreciate Referee #1's recognition and have carefully revised our manuscript according to these suggestions.

1) I do still quibble with the implication that it is only DMA that makes the SA dimer ion that is detected. Could be ion cluster fragmentation upon sampling. Furthermore, the supplemental states that the SA-DMA mechanism should go as SA to the 4th power, which is in opposition to their measurements.

Response:

We agree. DMA may not be the only stabilizer forming SA₂. We have added a discussion on the effects of other stabilizers (lines 252-257):

“Figure 4 shows that [SA₂]_{meas} is in accordance with [SA₂]_{sim} when considering the uncertainties. It means DMA, as a major stabilizer, can sufficiently explain the formation of SA₂. In addition to DMA, other gases could have participated in stabilizing SA₂. Kirby et al. (2011) reported the detection of SA₂ in the SA-NH₃ system of CLOUD experiments. Jen et al. (2014) found that methylamine (MA) and trimethylamine (TMA) contributed to the formation of neutral SA₂. Riccobono et al. (2014) observed that OOMs were able to be bounded to ion SA₂.”

Regarding the deviations between the measured data and the theoretical relationship, the complexity of atmospheres is likely to prevent new particle formation rates from conforming to theoretical expectations. Given that the influence of temperature on nucleation has been analyzed in the manuscript, we have use it as an example to provide further clarification (lines 136-139 in the Supplement):

“In actual observations, the measured data (Figure 1) did not fully conform to this functional relationship, which might arise from other influencing factors. For example, the substantial temperature variations likely accounted for the large fluctuations in particle formation rate observed at a given [SA₁] (Figure 6). If data points at TL (high temperature) are removed in Figure 6, it is not difficult to find that the rest of data points generally follow the functional relationship.”

2) At the warmer temperatures of this study, there will definitely be evaporation of some of the larger SA-DMA clusters. It is a mistake to consider the SA-DMA 1-1 cluster as the only temperature dependent factor for clusters. For example, 2-2 can quickly lose a DMA molecule.

Response:

Thanks. Our have improved this statement. In fact, we intended to describe that the formation of SA₁DMA₁ is a primary rate-limiting step for clustering, while the evaporation of other clusters remains non-negligible. We have removed the content that emphasizes on the decisive role of SA₁DMA₁ (formerly lines 293-295). We have also revised the description on the evaporation of SA-DMA clusters (lines 272-273 and lines 279-282, respectively):

“Compared with SA₂(DMA)₁₋₂, SA₁DMA₁ is more sensitive to temperature (Cai et al., 2022b). Thus, in addition to the evaporation of SA dimer, SA₁DMA₁ formation also acts as a rate-limiting step in clustering.”

“The observed reduction in the abundance of larger clusters at elevated temperatures is constrained by smaller ones (SA monomers and dimers), since the evaporation rates of SA trimers and tetramers show relatively low sensitivity to temperature (Olenius et al., 2017).”

3) I think some of the literature they cite (and other reports that they do not cite) indicates that ammonia synergism is important. This should be acknowledged in the conclusions and elsewhere: that it is probably not only SA-DMA but sufficient NH₃ must be present too.

Response:

We agree with the referee's comment. Moreover, like NH₃, OOMs synergism is also important to nucleation and need to be reflected in the manuscript, even if the supporting evidence is not yet sufficient. We have revised the manuscript in lines 221-228 and 355-359, respectively:

“Moreover, it is likely that OOMs and NH₃ played a synergistic role in nucleation under our campaign conditions. Multiple studies have detected clusters containing these precursors, such as SA-DMA-NH₃, SA-NH₃-OOMs, SA-NH₃ and SA-OOMs, in the presence of DMA in polluted atmospheres, and some of these clusters showed positive correlations with sub-3 nm particles (Yan et al., et al; Yin et al., 2021; Cai et al., 2024). In CLOUD studies, the contributions of OOMs and NH₃ to cluster formation were not as considerable as that of DMA (Kürten et al., 2018; Xiao et al., 2021). Given that our field measurements exhibited similar [NH₃] and relatively low [OOMs] compared to those in CLOUD experiments (Table 2), it follows that their effects were unlikely to be greater than DMA in our campaigns. Despite the potential participation of other precursors, the observation-simulation agreement suggests DMA is a major base that stabilizes SA clusters (Figure S5).”

“Based on the correlation between [SA₁] and particle formation rate, the identification of key clusters, and comparisons between simulations and measurements, we concluded that nucleating processes, mainly exemplified by SA₂ and J_{1,4}, could be largely attributed to SA-DMA collision. Other precursors, such as NH₃ and OOMs, might also have participated, since their concentrations were considerable and theoretically adequate to support nucleation.”

Referee #2:

1) Figure two caption can be modified to: Other species detected by CI-LToF-MS were not shown, because they are not “EXPECTED To PARTICIPATE IN” atmospheric nucleation. & The logarithm of values is annotated for “SOME” larger clusters.

2) Line 354: although other precursors, such as NH₃ and OOMs, might 'HAVE' also participated.

Response:

Thanks for Referee #2's specific suggestions regarding sentence phrasing. We have revised these sentences.

Editorial:

*1) The sections "Author contributions" and "Competing interests" must be included in *.pdf manuscript with the next revision.*

Response:

Revised as suggested.