Community Comment:

The manuscript by Shen et al. presents simulations of new particle formation from sulfuric acid (H_2SO_4) and dimethylamine (DMA) by molecular cluster kinetics modeling. The thorough comparisons of formation rates obtained with different thermochemistry input data sets and kinetic model assumptions provide very useful information on variations and uncertainties in predicted formation rates.

Response: We highly appreciate Dr. Olenius's attention to our study. We have carefully studied the comments, which are very helpful in improving the quality of our manuscript. We have provided detailed responses to each point of comment and made revisions in the manuscript accordingly.

I would like to bring up previous works applying ACDC-based particle formation rate look-up tables in large-scale 3D modeling, as the authors may not be aware of them (e.g. L118-119). These previous studies have applied look-up tables in the PMCAMx-UF, GEOS-ChemTOMAS and EC-Earth3 chemical transport or Earth system models with the following particle formation mechanisms:

- H_2SO_4–NH_3–H_2O with electrically neutral clusters (Baranizadeh et al., 2016; Croft et al., 2016),
- H_2SO_4–NH_3–H_2O + H_2SO_4–DMA with neutral clusters (Julin et al., 2018; Olin et al., 2022),

and

- H_2SO_4–NH_3 including both neutral and ionic species (Svenhag et al., 2024).

Response: We thank Dr. Olenius for providing these research summaries. We have revised the relevant sections in the revised manuscript to discuss and review the previous studies applying ACDC-based particle formation rate look-up tables in 3-D modeling in line 122-132 in the revised manuscript.

Most of these studies applied quantum chemistry data corresponding to the RICC2 method, as those were the only available complete data sets at the time. A comparison of global H_2SO_4–NH_3 particle formation and its effects as predicted by either DLPNO or RICC2 is presented by Svenhag et al. (2024). While DLPNO is the current best available method, it may underpredict formation rates under certain conditions (e.g. Besel et al., 2020), and the DLPNO-based rates were thus applied to assess the lower limits of the predicted effects.

Response: As mentioned above, although some studies have used ACDC-derived look-up tables in 3-D models for NPF simulations, the impact of input thermodynamic data, especially RI-CC2 and DLPNO on 3-D NPF simulation involving SA-DMA nucleation, is not yet clear. We have clarified the specific research gap addressed by our study in lines 132-138 in the revised manuscript.

We also agree that DLPNO may have uncertainties in fully accurately describing cluster thermodynamics, despite being currently recognized as the best quantum chemical calculation methods. We have clarified this in line 700-702 in the revised manuscript.

In the first studies, the calculation and interpolation of look-up tables were hard-coded for the given chemical components. The most recent work (Svenhag et al., 2024) applies automatized look-up table generator and interpolator that are applicable to arbitrary components (Yazgi and Olenius, 2023), enabling easy incorporation of tables obtained for different species and thermochemistry data. Automatization is needed especially for reading in and interpolating tables within the 3D model, as it is not feasible to maintain separate interpolation routines for different tables, corresponding to different chemical mechanisms and/or dimensions.

Response: We greatly appreciate the most recent works mentioned, which facilitate the integration of ACDC-based cluster dynamic simulations with 3-D modeling. For the single SA-DMA nucleation system focused in our study, the use of a hard-coded method is acceptable. However, if multiple nucleation mechanisms with different dimensions are simulated through look-up tables, the hard-coded method should be redundant. In such cases, the novel method of an automatized look-up table generator and interpolator would be much more feasible. We have added discussions on this topic in line 738-741 in the revised manuscript.

It can also be noted that the usage of pre-calculated formation rates (which is necessary in computationally heavy 3D models) involves simplifying assumptions on gas–particle kinetics, as there are no explicit interactions between the clusters and the nucleating vapors and larger nanoparticles. Therefore, a parameterization or look-up table approach may give biased results under some conditions even if the thermochemistry data were perfectly accurate. In computationally light-weight models, this can be circumvented by explicit simulation of the coupled gas–cluster–aerosol system (Olenius and Roldin, 2022), corresponding to a multicomponent adaptation of discrete–sectional modeling (Li and Cai, 2020).

Response: The potential impact of simplification of gas–particle kinetics using pre-calculated formation rates has also been concerned by reviewer #1. We have carefully studied the research conducted by Olenius and Roldin (2022) which represents the primary study concerning the explicit simulation of the coupled gas–cluster–aerosol system. We have examined the key dynamic processes, on which the usage of standard approach might exerts significant bias as demonstrated in this study.

Firstly, we evaluated the validity of the steady-state nucleation assumption by considering the system's e-folding time (time for clusters to reach (1-1/e) of their terminal concentration, following Li et al., (2023)). Specifically, we deemed the assumption reasonable if, under certain atmospheric conditions, the system's e-folding time is less than the simulation time step (300 s). As shown in Figure S14, results indicates that the e-folding time does not show a significant correlation with $J_{1.4}$. Under the majority of atmospheric conditions (77.3%), the nucleating system's e-folding time is less than 300 s. Instances where the e-folding time exceeds 300 s are primarily observed in winter clean conditions characterized by low temperature (T < ~270 K), low condensation sink (CS < ~0.003 s^{-1}), and low precursor concentrations (SA < ~10^6 cm^{-3}). These findings align with the observations of Olenius and Roldin (2022). It's important to emphasize that this e-folding time represents the duration required for the system to transition from having only precursor molecules to reaching near-equilibrium concentrations of various clusters. In reality, cluster concentrations generally do not start from zero. Therefore, the calculated e-folding time serves as an upper limit estimate. Given the predominance of atmospheric conditions where the e-folding time falls within or below the simulation time step of 300 s, consequently, the steady-state treatment is generally deemed reasonable for our WRF-Chem/R2D-VBS simulations.

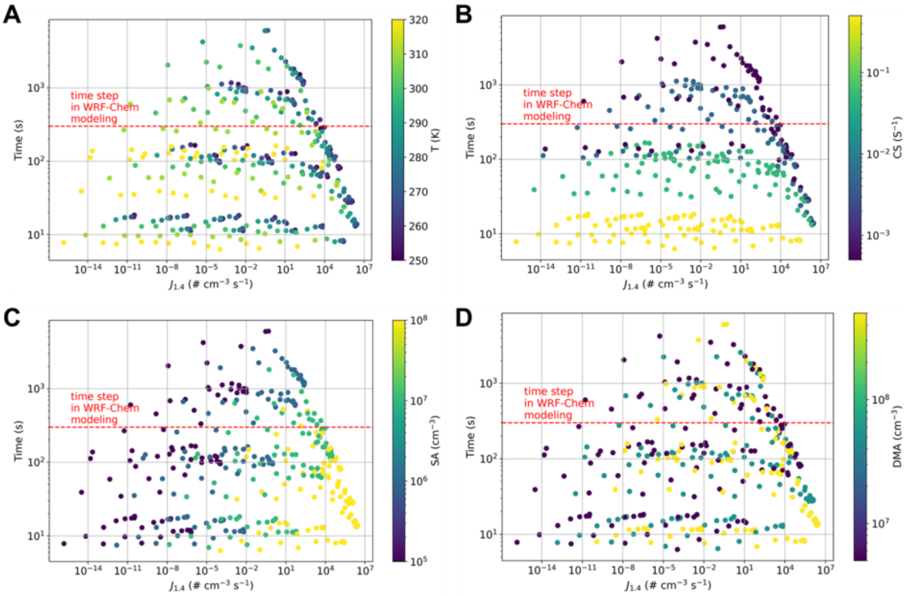

Figure S14. The variation of e-folding time with $J_{1.4}$ correlated with temperature (A), CS (B), SA concentration (C), and DMA concentration (D). The data points were calculated using a more sparse sequence of input parameters (T: 250, 260, 270, 280, 290, 300, 310, 320 (K); CS: 5.00×10^{-4}, 5.00×10^{-3}, 5.00×10^{-2}, 5.00×10^{-1} (s^{-1}); SA: 1.00×10^5, 1.00×10^6, 1.00×10^7, 1.00×10^8 (cm^{-3}); DMA: 5.00×10^6, 5.00×10^7, 5.00×10^8 (cm^{-3})) compared to those shown in Table S1.

We further investigated another common treatment that may introduce bias: neglecting cluster formation in consuming precursor during nucleation. Our examination focused on assessing the proportion of precursor consumption by cluster formation relative to precursor concentrations. As shown in Figure S15 and S16, we found that this proportion increases with $J_{1.4}$ for both SA and DMA. Under the majority of atmospheric conditions (82.0% for DMA and 57% for SA), proportions are below 10%. Proportions exceed 10% are predominantly observed in scenarios also characterized by low temperature (T < ~270 K) and low condensation sink (CS < ~0.003 s^{-1}), but with high deference in concentrations between DMA and SA. Specifically, elevated SA concentrations, which lead to significant DMA consumption through cluster formation, and vice versa, contribute to scenarios where precursor consumption by cluster formation exceeds 10%. It's noteworthy that our calculation of precursor consumption by cluster formation starts from zero cluster concentration. Also, in the real atmosphere, cluster concentrations are generally nonzero, leading to another upper limit estimate. Therefore, based on our analysis, it can be inferred that cluster formation may not introduce significant bias into NPF simulations under typical atmospheric conditions. We have added this additional analysis and discussion of the potential impacts of these common treatments in NPF simulations to line 717-731 in the revised manuscript and the supporting information.

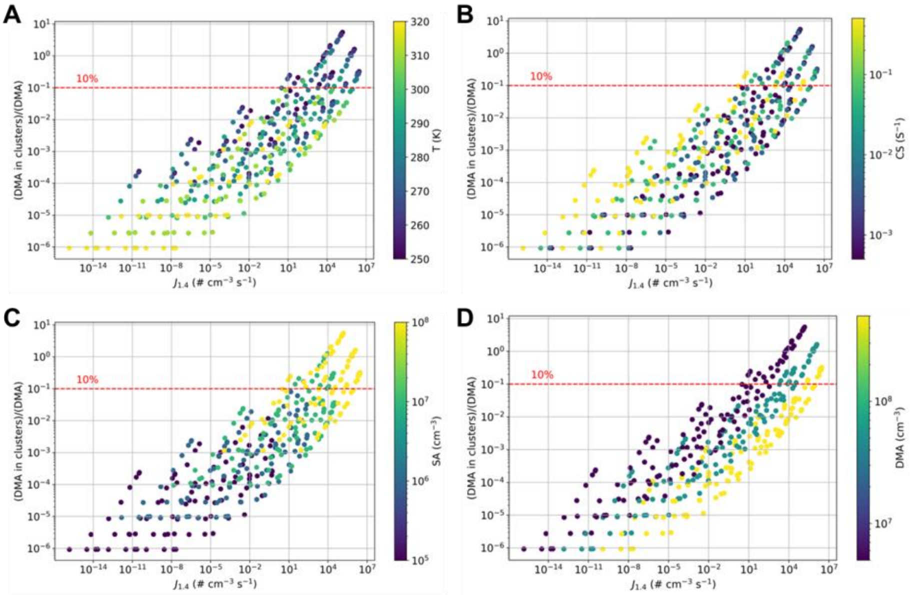

Figure S15. The variation of proportion of DMA consumption by cluster formation relative to precursor concentrations with $J_{1.4}$, correlated with temperature (A), CS (B), SA concentration (C), and DMA concentration (D). The input variables are consistent with Figure S14.

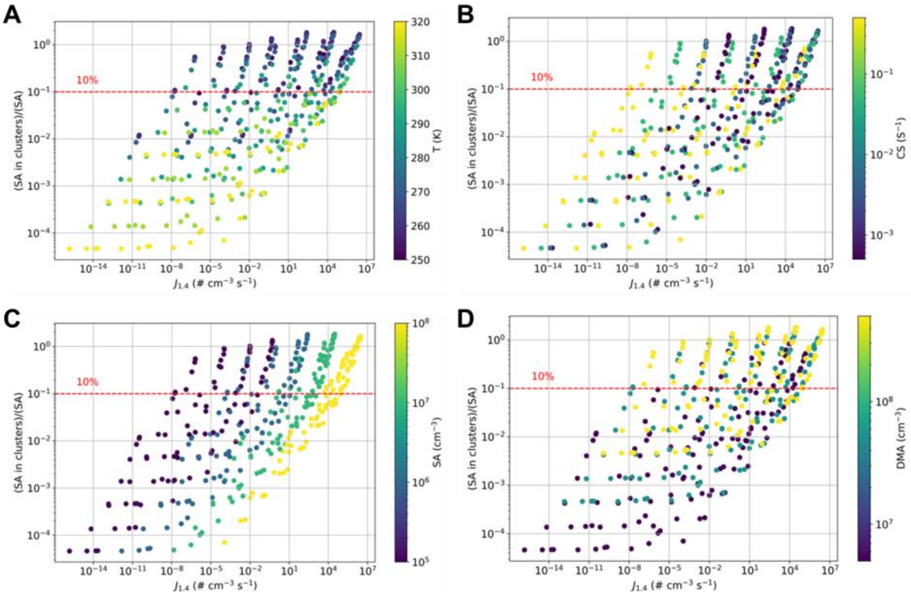

Figure S16. The variation of proportion of SA consumption by cluster formation relative to precursor concentrations with $J_{1.4}$, correlated with temperature (A), CS (B), SA concentration (C), and DMA concentration (D). The input variables are consistent with Figure S14.

Finally, I also encourage to refer to the ACDC code repository (https://github.com/tolenius/ACDC) in order to provide a reference for the model tools for reproducibility of simulation results.

Response: We have added the ACDC code repository in the revised manuscript in line 161.

REFERENCES

Li, Y., Shen, J., Zhao, B., Cai, R., Wang, S., Gao, Y., Shrivastava, M., Gao, D., Zheng, J., Kulmala, M., and Jiang, J.: A dynamic parameterization of sulfuric acid–dimethylamine nucleation and its application in three-dimensional modeling, Atmospheric Chemistry and Physics, 23, 8789-8804, 10.5194/acp-23-8789-2023, 2023.

Olenius, T. and Roldin, P.: Role of gas-molecular cluster-aerosol dynamics in atmospheric new-particle formation, Sci Rep, 12, 10135, 10.1038/s41598-022-14525-y, 2022.