Reviewer #2

General comments:

The work conducts comprehensive comparison of different cluster dynamics-based parameterizations for SA-DMA nucleation by integrating box-model simulations, 3-D modeling, and in-situ observations. It is found that ACDC_DB performs well in modeling 3-D NPF for both winter and summer in Beijing and shows promise for application in various atmospheric environments. Furthermore, ACDC_RM_SF0.5 exhibits effective applicability at ~280 K, but has limitations in predicting $J_{1.4}$ at elevated T. While Dynamic_Sim is applicable for simulating NPF in polluted atmospheres but makes significant overestimation of $J_{1.4}$ under conditions of high T and low CS.

The topic discussed in this paper is highly meaningful for developing parameterizations for various nucleation systems. The reported results are clearly presented and are relevant to the scope of Atmos. Chem. Phys. I recommend publication of this manuscript after consideration of the following comments.

Response: We thank the reviewer for recognizing and recommending our work.

Specific comments:

1) Lines 94-95: Please explain briefly the reason for considering such three simplifications within Dynamics_Sim.

Response: Generally, according to theoretical studies (Olenius et al., 2013, 2017; Ortega et al., 2012; Myllys et al., 2019), clusters $(SA)_1(DMA)_1$, $(SA)_1(DMA)_2$, $(SA)_2(DMA)_2$, $(SA)_3(DMA)_3$ and $(SA)_4(DMA)_4$ are considered the key clusters along the cluster formation pathways in SA-DMA nucleation. Under the polluted conditions (CS > ~1.0×10^{-2} s^{-1}), the evaporation rates of clusters $(SA)_1(DMA)_2$, $(SA)_2(DMA)_2$, $(SA)_3(DMA)_3$ and $(SA)_4(DMA)_4$ are negligible compared to their coagulation sink. Therefore, three simplifications are involved in derivation of Dynamic_Sim as described in line 93-102 in the revised manuscript. Details of the derivation of Dynamic_Sim can be seen in our previous study (Li et al. 2023).

2) Line 281: To make a clear understanding among readers, it would be better to provide the concept of the chemical initial and boundary conditions in WRF-Chem/R2D-VBS simulations.

Response: We appreciate the reviewer's suggestion which can help to enhance the readability of our manuscript. The chemical initial condition in WRF-Chem/R2D-VBS simulations refers to the concentration field of gas-phase/particulate chemical variables at the beginning of the simulation, standing as the evolution of these species before the simulation duration. The chemical boundary condition here refers to the fluxes or concentrations at the edges of the simulated domain (Brasseur et al. 2017). In WRF-Chem/R2D-VBS simulations, we use a 5-day spin-up to minimize the impact of chemical initial conditions on simulation results. Some explanations have been added in line 296-307 in the revised manuscript.

3) Figure 6C: It can be noted that ACDC_DB and Dynamic_Sim also exhibit an

underestimation of averaged PNSDs in the 2-100 nm range in comparison to observation. Can the authors account for the cause of this phenomenon?

Response: In fact, ACDC_DB and Dynamic_Sim do not exhibit a consistent underestimation of averaged PNSDs along the 2-100 nm range in comparison to observation. Similar to wintertime simulation, the PNSDs simulated by ACDC_DB and Dynamic_Sim for the summer season are relatively overestimated compared to observations in larger size range of 30-100 nm. This may not be evident due to overlapping curves in Figure 6C of the main text but is more noticeable in Figure A1 below.

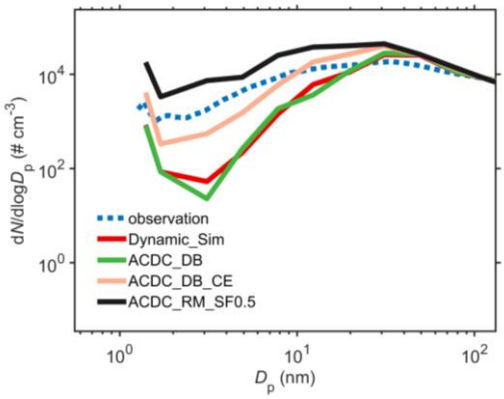

Figure A1. Comparison of observed and simulated PNSDs during August 18, 2019, to August 31, 2019, in Beijing. Simulations are conducted using parameterizations of Dynamic_Sim, ACDC_DB, ACDC_DB_CE, and ACDC_RM_SF0.5.

For the 2-100 nm range, we also compared the total number concentrations simulated from three main parameterizations and the ACDC_DB_CE with the observations (Figure S13). It can be noted that the number concentrations simulated by ACDC_DB and Dynamic_Sim are relatively consistent with the observations, whereas ACDC_DB_CE and another main parameterization ACDC_RM_SF0.5 tend to overestimate the number concentrations by a factor of 1.6 and 2.5, respectively. Combining the particle formation rates shown in Figure 6A for the three parameterizations, it can be concluded that the total concentrations of 2-100 nm particles are primarily influenced by nucleation. The discrepancies in PNSDs across different size ranges compared to the observations arise from the intrinsic treatment of growth processes in the 3-D model. We have added relevant clarifications in line 620-627 in the revised manuscript.

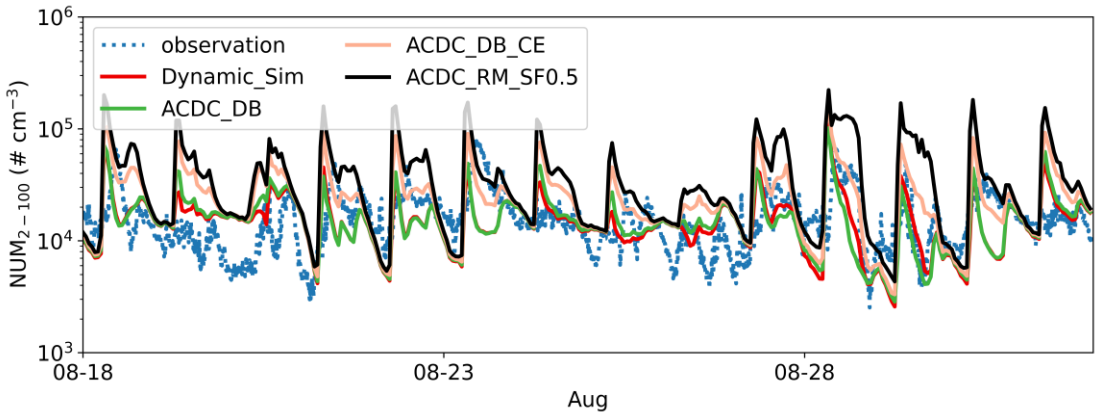

Figure S13. Comparison of observed and simulated aerosol number concentration within 2-100 nm during August 18, 2019, to August 31, 2019, in Beijing. Simulations are conducted using parameterizations of Dynamic_Sim, ACDC_DB, ACDC_DB_CE, and ACDC_RM_SF0.5.

Technical corrections:

1) Lines 143-144: "n and m represent the number of SA and DMA molecules in a cluster" should be "m and n represent the number of SA and DMA molecules in a cluster".

Response: The revisions have been made accordingly.

2) Lines 465 and 482: "ACDC_RM" should be "ACDC_RM_SF0.5".

Response: The revisions have been made accordingly.

3) Supporting Information, lines 37 and 38: "A: ΔG = 13.5 kcal/mol; B: ΔG = 12.9 kcal/mol" should be "A: ΔG = -13.5 kcal/mol; B: ΔG = -12.9 kcal/mol".

Response: The revisions have been made accordingly.

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