- **RC3**: 'Comment on acp-2022-487', Anonymous Referee #3, 13 Sep 2022

The authors thanks Referee #3 for the constructive comments, which have helped us to clarify and improve the manuscript. Below we address the comments, with the reviewer comments in black, and our response in blue. We have revised the manuscript accordingly. All changes made to the manuscript have been marked in the submitted Track-Changes version.

Very good work! The paper compares the nucleation process, between model results with two different nucleation schemes and CLOUD/ATom measurements, under the stratospheric condition. It's nice to see the authors relate the study to the SAI simulation, which could help to improve the SAI modeling accuracy. The overall reasoning in the paper is solid and well-justified.

We appreciate the positive comments and confirmation of the solid quality of this work.

I have some minor comments and corrections:

Line 81-83: Providing only two publication examples (i.e., Weisenstein et al., 2022, Laakso et al., 2022) seems not enough to prove that "BHN_V2002 has been used in most SAI modeling studies". It would be more convincing if the authors can tell us how many models use BHN_V2002. For example, there are many models involved in GeoMIP (Kravitz et al., 2013), it would be helpful if the authors can tell the GeoMIP community how many GeoMIP simulations use BHN_V2002 for nucleation simulation.

The two cited references are recent SAI model intercomparison papers. According to Kravitz et al. (2013), 13 out of 16 Geo-MIP models assumed prescribed or bulk stratospheric aerosol. The remaining 3 models generalized aerosols from SO_2 but no information on the nucleation schemes used was given in the paper. We have checked more SAI-related papers that explicitly consider size-resolved particle microphysics (including nucleation) and have revised the text regarding the BHN_V2002 scheme used in these studies.

"Indeed, the H_2SO_4–H_2O binary homogenous nucleation (BHN) parameterization developed two decades ago by Vehkamäki et al. (2002) (named BHN_V2002 thereafter) has been widely used in SAI modeling studies when nucleation process is explicitly considered (e.g., Tilmes et al., 2015; Jones et al., 2021; Weisenstein et al., 2022). Tilmes et al. (2015) described a Geoengineering Model Intercomparison Project (GeoMIP) experiment designed for climate and chemistry models, using the stratospheric aerosol distribution derived from the ECHAM5-HAM microphysical model (Stier et al., 2005) which calculated nucleation rates with the BHN_V2002 scheme. Both models (UKESM1 and CESM2-WACCM6) employed for a recent GeoMIP G6sulfur study (Jones et al., 2021) used the BHN_V2002 scheme. In another recent SAI study based on three interactive stratospheric aerosol microphysics models (Weisenstein et al., 2022), two models (MAECHAM5-HAM and SOCOL-AER) used BHN_V2002 scheme while the other (CESM2-WACCM) used an empirical nucleation scheme to calculate nucleation rate as a function of sulfuric acid concentration only (i.e, no dependence on temperature and relative humidity)."

Jones, A., Haywood, J. M., Jones, A. C., Tilmes, S., Kravitz, B., and Robock, A.: North Atlantic Oscillation response in GeoMIP experiments G6solar and G6sulfur: why detailed modelling is needed for understanding regional implications of solar radiation management, Atmos. Chem. Phys., 21, 1287–1304, https://doi.org/10.5194/acp-21-1287-2021, 2021.

Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and Petzold, A.: The aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys., 5, 1125–1156, https://doi.org/10.5194/acp-5-1125-2005, 2005.

Tilmes, S., Mills, M. J., Niemeier, U., Schmidt, H., Robock, A., Kravitz, B., Lamarque, J.-F., Pitari, G., and English, J. M.: A new Geoengineering Model Intercomparison Project (GeoMIP) experiment designed for climate and chemistry models, Geosci. Model Dev., 8, 43–49, https://doi.org/10.5194/gmd-8-43-2015, 2015.

Line 136: 4°x5° is too coarse. If possible, please repeat the simulations in 2°x2.5°. If not, the authors should discuss how much the grid resolution may influence the difference between the model results and observations, especially for the comparison between model results and ATom observations at one site in Figure 6.

We chose 4°x5° in this study because of computing cost constrain. It took about 2.5 days of wall clock time for one-year 4°x5° simulation and about 20 days of wall clock time for one-year 2°x2.5° simulation (more # of grid boxes plus shortened time step). Due to relative long lifetime of stratospheric aerosols, at least 1.5 years of spin-up time is generally required.

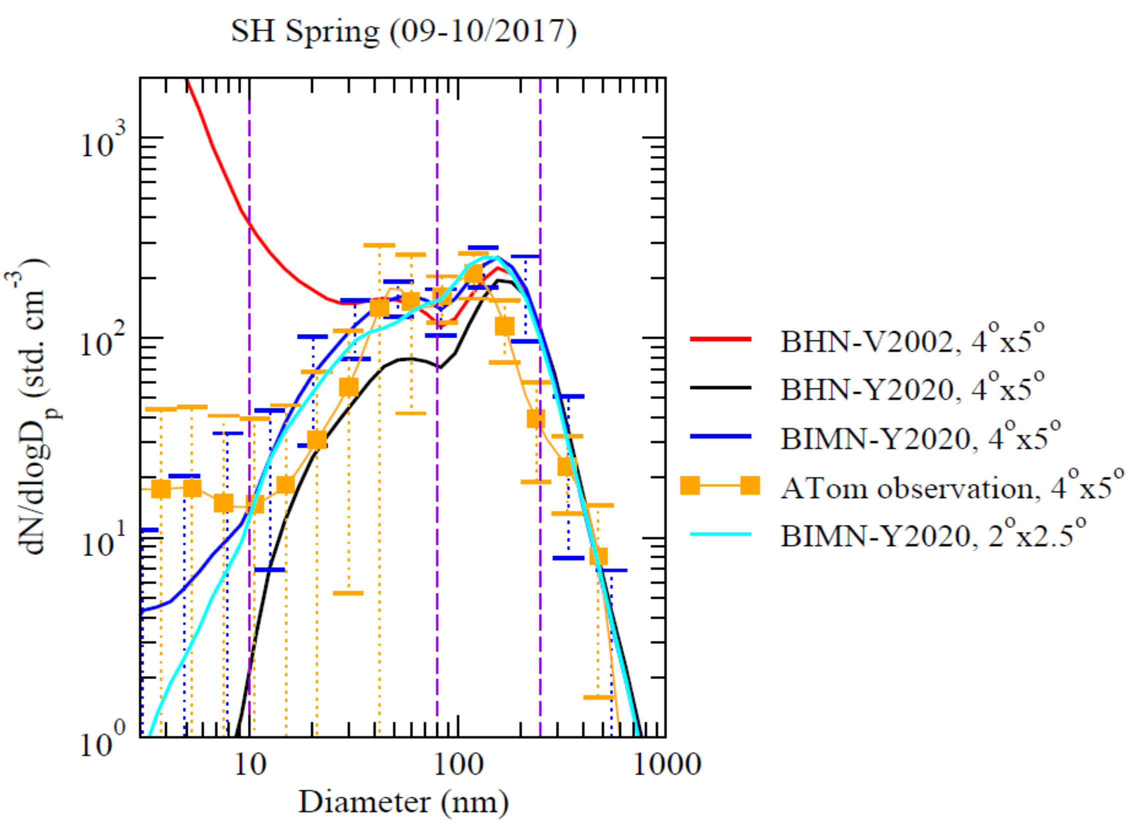

Figure R1. Same as Figure 6a in the main text but with a curve (cyan) added for BIMN-Y2020 case simulation at 2°x2.5° horizontal resolution.

While 4°x5° resolution is not ideal, it can be justified for the study of relatively homogeneous horizontal distributions of aerosols in the stratosphere. It should be noted that for comparisons shown in Fig. 6, the ATom data has been averaged to a 4°x5° gridbox (see Fig. 4). To check the effect of grid resolution, we run the BHN_Y2020 case at 2°x2.5° for two years (2016-2017). With the first 1.5 years as spin-up, we compare the modeling results with ATom 3 (09-10/2017, SH Spring) in Figure R1 (corresponding to Fig. 6a). It can be seen that the difference is generally small compared to the variations of the measurements and model simulations (as indicated by the error bars) and thus will not affect the main conclusions of this paper. Please note that at least some of the difference between the blue (4°x5°) and cyan (2°x2.5°) curves is caused by the different areas represented by the two curves (one for a 4°x5° grid box and the other for a 2°x2.5° grid box).

Line 274: why the tropics are selected as "(0°S-30°S)" instead of "(30°S-30°N)"?

Thank you for pointing this out. It was a typo. Corrected.

Line 296: Figure 3 needs to be optimized:

(1) Set shared x or y axis label among figures (a) to (f).

y-axis label is now shared. We keep x-axis label for each panel for clarity.

(2) Adjust the location/size of figures (g) and (h).

Slightly adjusted.

(3) There is a horizontal dashed line on the top of the figure (h), which should be deleted.

Fixed.

Line 296: for Figure 3 (g),

(1) is it a coincidence that three solid lines end up with a similar nucleation rate (about 0.02 std. cm^{-3} s^{-1}) at approximately 17.5 km?

This is a coincidence for 30S-30N average. As can be seen from Fig. 3a-3c at ~17.5 km, there exist variations in the latitude direction although the 30S-30N average is about the same.

(2) why there is an elbow point (at around 20 km) in the red solid line? In another word, why does the nucleation rate from BHN_V2002 has a much larger changing rate with height above 20 km, compared to below 20 km?

This is a good observation. It is caused by much smaller vertical gradient in BHN_V2002 nucleation rates within ~ 17-20 km (see Fig. 3a), likely a result of different dependences of nucleation rates based on different schemes on *T*, RH, and [H_2SO_4] which have large vertical variations (see Fig. 2). We have pointed this out in the revised text.

Line 319: "BHNV_2002" should be "BHN_V2002".

Corrected.

Line 390-391: I think that the competition between nucleation and condensation mentioned by Laakso et al. (2022) might be a complement to the "nonlinear process" (Line 390-391) mentioned by the authors.

Yes. We added a sentence to point out the work of Laakso et al. (2022) with regard to the competition: "The competition between nucleation and condensation for available sulfuric acid gas has been shown to be important for SAI studies (Laakso et al., 2022)."

Line 409: I don't understand the sentence: "Finally, the observed PNSDs show a clear AccuM2 in all seasons except Fall but the model does not predict the existence of the mode at all."

Based on Figure 6, the model may underpredict the AccuM2, especially in summer. But we cannot say "the model does not predict the existence of the AccuM2 mode at all".

What's more, the authors say "the model-simulated AccuM2 standard deviations are larger in SH Winter and Spring but are smaller in SH Summer and Fall" in Line 505. If "the model does not predict the existence of the AccuM2 mode at all", there would be no "model-simulated AccuM2 standard deviations".

That's a good and valid point. "underpredict" is a more accurate word for this. We have modified the sentence to reflect this.

"Finally, the observed PNSDs show a clear AccuM2 in all seasons except Fall but such a mode cannot be clearly seen in the model simulated PNSDs, indicating that the model underpredicts the concentrations of AccuM2 mode particles."

Line 437: The citation (Clement and Harrison, 1992) is missed in the References. Please check and make sure all the citations in the main text are correspondingly listed in the References.

Thanks for noticing this. We have added the references and double-checked other citations.

Line 475: Suggest changing "SAI efficiency" to "SAI radiative efficacy". Radiative efficacy refers to the radiative forcing normalized by the aerosol injection rate, which is widely used in SAI studies (e.g., Dai et al., 2018).

Modified as suggested: "Recent studies indicate the dependence of SAI radiative efficacy (Dai et al., 2018) onthe particle size distribution (NASEM, 2021) …"

In the discussion part, I think the authors can highlight the importance of model development for reducing model uncertainties of SAI simulations. Some other SAI-related model development work (e.g., Golja et al., 2021, Sun et al., 2022) is worth mentioning.

Yes, we have added the following sentence in the discussion part: "The present work highlights the importance of advancing scientific understanding of processes controlling properties of stratospheric particles as well as further development, improvement, and validation of models for reducing uncertainties of SAI simulations (e.g., Golja et al., 2021, Sun et al., 2022)."

For the next step, I hope the authors could consider comparing the modeled aerosol radiative forcing based on the two different nucleation schemes, which could help the Solar Geoengineering community to have a clear feeling about how much can different nucleation schemes influence the SAI radiative efficacy.

Thanks for the suggestion. That's our plan for the near future.

The papers mentioned above:

Dai et al., 2018: https://doi.org/10.1002/2017GL076472

Golja et al., 2021: https://doi.org/10.1029/2020JD033438

Kravitz et al., 2013: https://doi.org/10.1002/2013JD020569

Laakso et al., 2022: https://doi.org/10.5194/acp-22-93-2022

Sun et al., 2022: https://doi.org/10.1029/2021MS002816