- **RC1**: 'Comment on acp-2022-487', Anonymous Referee #1, 26 Aug 2022

The authors thanks Referee #1 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.

Manuscript by Fangqun et al studies nucleation in the stratospheric condition by evaluating different nucleation schemes against CLOUD laboratory measurements and ATom observations in the southern hemisphere stratosphere. Study showed that in the recently developed kinetic nucleation model nucleation rate is much better in agreement with measurements compared to widely used binary homogeneous nucleation. This paper is exactly the type of research that modelers of stratospheric aerosols need, especially if you are studying stratospheric aerosol injections. Good indicator for excellent study is that after reading the paper, you want to take something from it to your own research. In this case it is the nucleation scheme.

Manuscript is well written, and when questions came to my mind, those were answered in the following lines. Or at least as well as possible. Overall this is an interesting and excellent study and it is difficult to find any major or even minor issues. I have just few very minor comments which would clarify some specific points:

We appreciate the positive comments and confirmation of the importance of this work. Please see below for our point-to-point replies and clarifications.

Model and data section:

It would be interesting to know how competition between nucleation and condensation for sulfuric acid vapor is done in GC-APM.

In GC-APM, nucleation is calculated before condensation using a time-splitting technique. Therefore, no competition between nucleation and condensation for sulfuric acid vapor is considered. In most conditions, nucleation consumes only a very small fraction (<1%) of sulfuric acid vapor in the air and the time splitting does not affect the results. When nucleation rate is high, reduced time step for nucleation and growth is used to ensure that the fraction of sulfuric acid vapor consumed by nucleation each time step is small. The GC-APM uses a semi-implicit scheme to calculate sulfuric acid condensation together with sulfuric acid gas phase production to ensure that the change of sulfuric acid vapor concentration is smooth. We have clarified this in the revised text.

P6 L206, After reading the abstract and introduction I got the impression that in this paper only the BHN of Vehkamäki and new BIHM scheme are evaluated. This line is the first time where BHN of Yu et al. (2020) is mentioned. I think it would be good to somehow introduce this nucleation scheme earlier (just by 1 or few line(s)) and say with just few words how it differs from the BHN of Vehkamäki (new look up tables(?) I assume).

Modified as suggested:

"Figure 1 compares nucleation rates based on the following three different schemes with CLOUD measurements under stratospheric temperature range (*T* = 205–223 K): BHN of Vehkamäki et al. (2002) (BHN_V2002), BHN of Yu et al. (2020) (BHN_Y2020), and BIMN of Yu et al. (2020) (BIMN_Y2020). BHN_V2002 and BHN_Y2020 differ in term of thermodynamic data and nucleation approach used (Yu et al., 2020)."

P6 Figure 1 (and later figures): Really minor thing but it would be more clear if (a) and (b) were upper left of the panel and not in the title and that order would be a-b-c in upper panels and not a-c-e as in figs 2-3.

Modified as suggested.

P7 L246 and later in the text, at least I am not familiar with "std" in units. Could this be opened up?

Here "std. cm^{-3}" refers to per cubic centimeter at standard temperature and pressure, 273 K and 1013 hPa, respectively. This has now been specified when ""std. cm^{-3}" is firstly used.

P7 L251 and L253, brackets in [H2SO4]

Brackets in [H_2SO_4] refer to H_2SO_4 vapor concentrations. This is now pointed out when [H_2SO_4] is first used.

P11 L349 You could clarify that here you refer to measurements even though it is obvious after reading the next couple of lines.

Modified as suggested.

P12 L361-368 I have been struggling with this and I am not sure if the standard deviation in the largest model is surprisingly large or is it even large or not. Number concentration in AccuM2 is much lower compared to AccuM1 and Aitken modes and thus in the linear scale the size range of the standard deviation bar is not actually large as it seems to be in logarithmic scale.

You are correct -- the absolute values of the standard deviation in AccuM2 are actually smaller than those in AccuM1 and Aitken modes. What we meant in the discussion is the normalized standard deviation (i.e., the standard deviation divided by the mean) that shows the relative variations. We have clarified this in the revised text.

Figures 5 and 6 and captions: There abbreviation of particle number size distribution is "PSD" while in the text it is "PNSD"

Good catch. All "PSD" have been changed to "PNSD" for consistency.

P13 L389-391 I think this is kind of expected as there is lower nucleation rate in BIMN compared to BHN_V2002, there is more available sulfuric acid vapor for condensation. However, the BHN-V2020 line is lower than BIMN and BHN_V2002 regardless of the size of the aerosol, which is interesting. It means that particulate sulfate (+small amounts of some other species) burden is lower in BHN-V2020 compared to others. Where is this “missing” sulfate in BHN-Y2020? In the gas phase, or in some other location, or removed from the atmosphere? If you could easily give an answer to this, it would be interesting to know.

Actually, there is no “missing” sulfate in BHN-Y2020. While the line of BHN-Y2020 is lower than that of BIMN and BHN-V2002 for particles of smaller sizes (<~ 300 nm), it is slightly higher for larger particles (>~ 300 nm). In PNSD plots, these slightly higher values for BHN-Y2020 for larger particles cannot be clearly seen in the PNSD plots but can be seen in the values given in the data files or mass size distributions. This is consistent with the competition of sulfuric acid gas between pre-existing larger particles and nucleated smaller particles. This has been clarified in the revised text.

P13 L408 I find this bi-mode structure of accumulation size region really interesting. What do you think, should this be taken into account in modal aerosol schemes especially in stratospheric conditions and add one extra mode between usually used accumulation and coarse mode? Your results are not the only case which would speak for it. Of course then in the modal scheme bi-mode structure would be artificial, as it still remains unknown what is causing it.

More research is needed to characterize and understand this bi-mode structure. If it is indeed a common and wide-spread feature, it should be taken into account both in modal and sectional aerosol schemes. We agree that it is critical to understand what is causing it.

P14 L439 Is there bi-mode structure of accumulation mode in a volcano perturbed atmosphere? I would say that there is the coarse mode and one accumulation mode.

It depends on how we define coarse mode sizes. In the aerosol research community, coarse mode refers to particles generally larger than 2.5 μm (i.e, PM2.5, PM10, etc.). In this case, we can still say that there is a bi-mode structure of accumulation mode in a volcano perturbed atmosphere.