- **RC2**: 'Comment on acp-2022-487', Anonymous Referee #2, 28 Aug 2022

The authors thanks Referee #2 for the thoughtful 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.

This is really interesting. I like the combination of modeling, field experiment measurements, and chamber measurements. Getting a chamber to cooperate with stratospheric conditions is no small feat. There are some important gaps in the study that I'd like to see resolved, mainly having to do with the applicability of your datasets.

More specifically, you show that these new nucleation schemes better match observations. But your observations do not match what is usually thought of as hypothetical SAI conditions. This introduces a potential source of error in your study that is not well discussed.

We appreciate the positive comments about this work. Please see below for our point-to-point replies and clarifications about the gaps.

Comments:

I'd like to see you discuss volcanic eruptions more. Presumably if you're coming up with new assessments of past modeling of SAI it would also affect past modeling of volcanoes. Did we miss something very important in our previous assessments of volcanic aerosol microphysics? Did that affect our estimates of radiative forcing or chemistry?

Detailed measurements are needed to properly assess the modeling performance. This study focuses on the period where in-situ ATom airborne measurements are available. As the reviewer pointed out in the next comment, the ATom measurement period does not have a high stratospheric loading. It remains to be investigated if previous assessments of volcanic aerosol microphysics missed something important. We expect the uncertainties in the nucleation schemes and unknown cause of the bi-modal structure of accumulation mode particles will affect particle optical properties and surface area and thus radiative forcing or chemistry. The exact effects remain to be studied, ideally with good in-situ particle size distribution measurements such as those from ATom. We have added some discussions on this in Section 4.

The period chosen (which overlaps with ATom) doesn't have a high stratospheric loading, and the particle size is substantially smaller than would be experienced under SAI. Is there any reason to think that microphysical behavior will be different under SAI conditions (or volcanic conditions)? This is exemplified in Figure 4 – while it's clear that the updated schemes better match observed CN3 than the 2002 scheme, this is only for a narrow range of CN3 and is poorly constrained for higher CN3 numbers.

We agree that the particle size during the ATom period is substantially smaller than would be experienced under SAI. The stratospheric particle properties during the ATom period can be considered to be those of background stratosphere. We expect that microphysical behavior

described in this work will be similar under SAI conditions (or volcanic conditions). However, the effect under these conditions remains to be investigated. We agree that Figure 4, representing the background LMS, is only for a narrow range of CN3. Unfortunately, we do not have measurements for higher CN3 numbers that can be used to constrain the model.

12 km isn't very high in altitude – that won't reach the stratosphere in many places, so the fact that your scheme better matches observations doesn't necessarily show that it better matches observations in the stratosphere. I would like to see more discussion on how this limitation affects your conclusions about stratospheric NPF. You discuss some of this in Section 3.2, but I'm having trouble interpreting the applicability and limitations of your study. Relatedly, on lines 233-234, which volcanic event and how much SO2?

Yes, this is a limitation of current measurements – ATom can only reach up to ~ 12 km altitude. Similar measurements (i.e., particle size distributions down to ~ 3 nm) at higher altitudes will be needed to evaluate nucleation schemes at higher altitudes. It should be noted that, in addition to using ATom data, in this work we also use the CLOUD laboratory measurements to assess the nucleation schemes (Fig. 1).

The volcano mentioned on lines 233-234 is the eruption of the Bezymianny volcano (55.98˚N, 160.59˚E) on December 20, 2017. The amount of SO_2 injected to the stratosphere (up to 18 km) due to this volcanic was $5x10^6$ kg S according to the volcano emission inventory (Carn et al., 2015). We have added the information of this volcano in the revised text.

Carn, S. A., Yang, K., Prata, A. J. and Krotkov, N. A.: Extending the long-term record of volcanic SO2 emissions with the Ozone Mapping and Profiler Suite nadir mapper. Geophys. Res. Lett., 42: 925– 932. doi: 10.1002/2014GL062437, 2015.

I'd like to see more description about the chamber. There is more to the stratosphere than just cold temperature – one needs to include low pressure, harsh radiation, composition, etc. Are you actually reproducing stratospheric conditions or just stratospheric temperatures? And if the latter, how relevant are your conclusions for stratospheric NPF?

Just the stratospheric temperatures. NPF in the stratosphere is generally considered to be involving H_2SO_4-H_2O. The physics underlying the H_2SO_4-H_2O nucleation is generally well understood although there is uncertainty in the thermodynamic data of pre-nucleation clusters. Presently we do not have theoretical and experimental evidence indicating the effect of low pressure and harsh radiation. The effect of H_2SO_4-H_2O binary composition is taken into account by the present nucleation schemes.

Figures 2, 3, and 5: I don't have a good sense for which scheme gives you better answers. What are these "supposed to" look like?

These figures show us the large difference caused by different nucleation schemes. Unfortunately, we do not have measurements to tell us what these are "supposed to" look like. Nevertheless, measurements given in Figures 1, 4, and 6, although limited, did provide some constrains on what these are supposed to look like under the conditions specified.

You make a good case for a second accumulation mode. But there are many schemes (both modal and sectional) that take a second accumulation mode into account. Perhaps they don't get the processes correct that would create such a mode, but they do have it. It might be useful to point out what those schemes are doing wrong.

We were not able to locate specific references showing "many schemes (both modal and sectional) that take a second accumulation mode into account". While some modal schemes use different modes (like MAM) to represent particles of different sources and sizes, we did not find any model to specifically separate accumulation mode particles in the stratosphere into two modes.

You could do a bit more work (or some discussion) to characterize your uncertainty. On lines 392-431 you discuss several sources of potential error, including missing processes or uncertainty in nucleation rates. Do you have a sense as to whether these sources are dominant or secondary? If the former, your results are at the risk of being made obsolete by someone who addresses those other sources of error.

It is hard to robustly quantify various uncertainties for the reasons pointed out in the main text. A certain source could be dominant in certain aspect (for example, the cause of the second accumulation mode) while others could be secondary. We raised these issues in the study so that the research community are aware of these. We will be happy to see these sources of errors being addressed in future research toward advancing our understanding of processes controlling size distributions of stratospheric aerosols.