Atmospheric Chemistry and Physics Discussions

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Comment on acp-2022-487

Anonymous Referee #2

Referee comment on "Particle number concentrations and size distributions in the stratosphere: implications of nucleation mechanisms and particle microphysics" by Fangqun Yu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-487-RC2, 2022

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.

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?

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.

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?

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?

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?

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.

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.