

Dear Andreas,

Thank you very much for obtaining the review comments of our manuscript and your suggestion. Below is my reply to the reviewer's comments and your suggestion (in blue).

The reviewer responded to me with the following comment:

"The authors have done a good job of pointing out that the questions I asked are really hard and probably can't be answered in one paper. But that also means that their paper probably shouldn't be so conclusive. I'd like to see them take more of the tack that they mention in their reply - pointing out shortcomings that the research community needs to pay attention to in the coming years. I think a paper along the lines of "There's a lot we don't know, and it's really important for us to find out" would be more useful than a paper that limits itself to narrow descriptions of the results of their tools. I think this is more of a comment about framing than anything - the authors don't need to come up with new results to address my comments."

To me, it seems that this comment can be taken up in the summary and discussion section. Please let me know how you want to approach the comment by the reviewer.

I agree with the reviewer about the importance of "*pointing out the shortcomings that the research community needs to pay attention to in the coming years*". Actually this is the main focus of this manuscript by pointing out (and showing) the limitation of the nucleation scheme (i.e., BHN_V2002) widely used in the community and the unknown cause of the bi-modal structure of accumulation mode particles. I also agree that "*There's a lot we don't know, and it's really important for us to find out*". To take your suggestion, I have added a sentence in the Summary and Discussion section to emphasize these points. I have also slightly modified the last sentence of the abstract to reflect this. The changes are marked in red below.

Summary and Discussion

... 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. **In addition to what we have shown in this study, there are likely other uncertainties or missing processes we do not know and the community needs to identify and resolve these.** The present work highlights the importance of advancing scientific understanding of processes controlling properties of stratospheric particles, **identifying important processes that the present models might have missed**, and further development, improvement, and validation of models for reducing uncertainties of SAI simulations (e.g., Golja et al., 2021, Sun et al., 2022).

Abstract:

... Considering the importance of accurate PNSDs for projecting realistic radiation forcing response to stratospheric aerosol injection (SAI), it is essential to understand and incorporate such potentially important processes in SAI model simulations **and carry out further research to find out what other processes that the present models might have missed.**

Please let me know if you have any questions.

Best regards,

Fangqun