Dear Dr. Boucher,

Thank you very much for your comment, which we very appreciate. We address all your points raised in blue color below.

Best,

Andrea & Sandro & Co-Authors

The authors are right that the numerical aspects of the aerosol microphysical scheme should not be overlooked. In the S3A model (Kleinschmitt et al., 2017), we opted for an adaptive sub-timestepping approach as a compromise between accuracy and computation cost (see section 2.2.5 of the reference below for a full description). Here is an extract of our study without the equation:

"As both processes, nucleation and condensation, consume H_2SO_4 vapour while having very different effects on the particle size distribution, the competition between the two processes has to be handled carefully in a numerical model. Furthermore, this has to be done at an affordable numerical cost, as we aim to perform long global simulations. We address this in the S3A module using an adaptive sub-timestepping. After computing the H_2SO_4 fluxes due to nucleation and condensation in kg H_2SO_4 s^{-1} from the initial H_2SO_4 mixing ratio, a sub-timestep, Δt_1, is computed such that the sum of both the nucleation and condensation fluxes consumes no more than 25 % of the available ambient H2SO4 vapour... This sub-timestepping procedure is repeated up to four times ... The fourth and final sub-timestep is chosen so that the sum of all sub-timesteps is equal to one timestep of the model atmospheric physics. This joint treatment of nucleation and condensation is imperfect, but it has the advantage of being much more computationally efficient than the usual solutions consisting of taking very short timesteps and much simpler than a simultaneous solving of nucleation and coagulation. The number of sub-timesteps could be increased for increased numerical accuracy; however, a number of four sub-timesteps was considered to be sufficient."

You may want to benchmark this approach (using different numbers of sub-timesteps) against yours in terms of accuracy and computational cost.

Reference

Kleinschmitt, C., Boucher, O., Bekki, S., Lott, F., and Platt, U.: The Sectional Stratospheric Sulfate Aerosol module (S3A-v1) within the LMDZ general circulation model: description and evaluation against stratospheric aerosol observations, Geosci. Model Dev., 10, 3359–3378, https://doi.org/10.5194/gmd-10-3359-2017, 2017.

Thank you very much for your comment and for pointing to Kleinschmitt et al. (2017). We had a look at that paper. We think that the method which is presented there is subject to similar problems described in this paper when exposed to continuously large H_2SO_4-supersaturation as they appear in a continuous SO_2 injection scenario. Same as our model, S3A works fine under background conditions and for the representation of volcanic eruptions. SOCOL-AER is also able to reproduce background sulfuric acid aerosol concentrations in the stratosphere under background conditions (e.g., Feinberg et al., 2019) as well as under conditions of volcanic eruptions (e.g., Sukhodolov et al., 2018, Quaglia et al., 2023). However, when exposed to continuously large H_2SO_4-supersaturations under conditions of continuous SO_2 injections the numerical solution of the semi-implicit scheme gets numerically unstable.

From the description in Kleinschmitt et al., 2017, it is not clear which H_2SO_4-supersaturation is used to calculate the nucleation rate:

“After computing the H2SO4 fluxes due to nucleation and condensation in kg H2SO4 s−1 from the initial H2SO4 mixing ratio, a sub-timestep, is computed such that the sum of both the nucleation and condensation fluxes consumes no more than 25 % of the available ambient H2SO4 vapour.”

If understood correctly, Kleinschmitt et al. (2017) start with a large H2SO4-supersaturation resulting from the H2SO4-production of the previous 30 min dynamical timestep (i.e. $H2SO4_0$ in Kleinschmitt et al., 2017), which needs to be balanced by condensation and nucleation. However, using this initially very large H2SO4 supersaturation for the calculation of nucleation rates leads to significant overestimation of nucleation mass fluxes at large H2SO4 supersaturations. Please have a look at chapter S2 in the supplement of this manuscript. We show that it is important to properly distribute the H2SO4 production over the microphysical sub-loops as well. Just continuously updating the initially large $H2SO4_0$ concentrations after each sub-loop resulted in non-physical features in the resulting aerosol size distribution under high H2SO4 supersaturations.

Spitting the 30-min-time step into 4 parts proportional to 25% of the total H2SO4 nucleation and condensation mass flux probably results in timstep lents of t1<t2<t3<t4, where the first one is the shortest, due to higher supersaturations in the beginning and thus stronger condensation and especially nucleation rates. However, making the sub-loops proportional to mass flux is probably also subject to significant biases under larger H2SO4 supersaturations, since the nucleation flux is only a tiny fraction (about 1%) of the condensation flux when applying a very short microphysical time step (see our Table S1 in the supplementary material). Having a too large microphysical timestep in combination with larger H2SO4 supersaturations results in a strong overestimation of the nucleation rate.

The parameterisation presented in Kleinschmitt et al., 2017 might work for small H2SO4 supersaturations or short perturbations (e.g. Volcanic eruptions), but we suspect that the solution presented is subject to similar problems as we present in our manuscript. Maybe it might make sense to sensitivity test your model to conditions of continuously high H2SO4 supersaturations (i.e. SAI conditions).

Andrea & Sandro & Co-Authors

References:

Feinberg, A., T. Sukhodolov, B. P. Luo, E. Rozanov, L. H. E. Winkel, T. Peter, and A. Stenke (2019): Improved tropospheric and stratospheric sulfur cycle in the aerosol-chemistry climate model SOCOL-AERv2, Geosci. Model Dev., DOI:10.5194/gmd-2019-138.

Kleinschmitt, C., Boucher, O., Bekki, S., Lott, F., and Platt, U.: The Sectional Stratospheric Sulfate Aerosol module (S3A-v1) within the LMDZ general circulation model: description and evaluation against stratospheric aerosol observations, Geosci. Model Dev., 10, 3359–3378, https://doi.org/10.5194/gmd-10-3359-2017, 2017.

Quaglia, I., Timmreck, C., Niemeier, U., Visioni, D., Pitari, G., Brodowsky, C., Brühl, C., Dhomse, S. S., Franke, H., Laakso, A., Mann, G. W., Rozanov, E., and Sukhodolov, T.: Interactive stratospheric aerosol models’ response to different amounts and altitudes of SO2 injection during the 1991 Pinatubo eruption, Atmospheric Chemistry and Physics, 23, 921–948, https://doi.org/10.5194/acp-23-921-2023, 2023.

Sukhodolov, T., Sheng, J.-X., Feinberg, A., Luo, B.-P., Peter, T., Revell, L., Stenke, A., Weisenstein, D. K., and Rozanov, E.: Stratospheric aerosol evolution after Pinatubo simulated with a coupled size-resolved aerosol–chemistry–climate model, SOCOL-AERv1.0, Geosci. Model Dev., 11, 2633–2647, https://doi.org/10.5194/gmd-11-2633-2018, 2018.