Dear Dr. Niemeier,

Thank you very much for your review, which helped us improve the manuscript. We appreciate the time you invested and address all your comments in blue color below.

Best,

Andrea & Sandro & Co-Authors

The paper by Vattioni et al on the different handling of the operator splitting of nucleation and condensation and, sub-timestepping handles an important aspect of uncertainties in the evolution of sulfate aerosols in the stratosphere. The paper is well written and needs minor revisions.

My main comments are related to the last section. My recommendation is to discuss your work in more detail in relation to Wan et al (2013,2023), not just to the schemes in HAM 1 and HAM2, but also in relation to their method that solves production, condensation and nucleation simultaneously. You see, somehow, similar problems, and their article provides a solution. It may be difficult to upgrade your model, but you should discuss the conclusion that it would be better to solve multiple processes simultaneously. It may be good to add a figure of the nucleation rates as well.

We have addressed this in the section from 407 onward. We have added the following two sentences: “However, the best way forward would be to directly implement an implicit solver of H2SO4 production, condensation and nucleation into the next version of SOCOL as it is presented in Wan et al. (2013). This will likely generate more accurate results while avoiding the need for sub-loops, which are computationally more expensive. However, these implicit solvers have not yet been tested for numerical stability under conditions of continuously larger SO2 injections as they would occur in climate intervention scenarios.”

We think the nucleation rates provided in Table S1 in the supplement are enough. An additional figure for nucleation rates only would be beyond the scope of the paper.

Can you give a recommendation how to proceed? What is the best option for simulations of SAI or volcanic eruptions? Will this also apply to other models?

An outlook is given starting from lines 462 onward. There is no best “option” for simulation of SAI or volcanic eruptions. The microphysical settings are very sensitive to the model resolution (horizontal, vertical and temporal) as well as to the specific injection/emission scenario applied (location and injection rate and time). It is unclear if the numerical instability under large H2SO4-supersaturations also applies to other models, however, it is definitely something other modeling groups should be aware of. This is discussed in the indicated paragraph. We have added the following sentence as a recommendation how to proceed:

“To increase confidence about different aerosol microphysics modules incorporated in the various aerosol-chemistry climate models we recommend conducting a model intercomparison study, which focuses on numerical stability of aerosol microphysics under conditions of large H2SO4 supersaturation.”

Linee 7 and 10: 'of of'

Thanks for spotting, we corrected this.

Line 38: The aerosol scatter

We changed to “scatter”

Line 140: How do you handle other processes, e.g. sedimentation? A long timestep (2h) may reduce sedimentation artificially in case the aerosols sediment into the next gridbox only.

The simple updraft scheme used for sedimentation in the original 2-D AER code was replaced by the numerical scheme of Walcek et al. (2000) in Feinberg et al. (2019). This reduced numerical diffusion and improved mass conservation.

The sedimentation speed of a 320 nm 70wt%-H_2SO_4 aerosol is about 1 meter per 2 hours at 50 hPa in the stratosphere. For smaller particles it is even less. Therefore, calling the sedimentation routine every two hours is fine. We do not expect artificial reduction or increase of sedimentation.

2.2 Socolv4: Changing the resolution of the model changes transport (e.g. Niemeier et al, 2020). This should be kept in mind.

Yes, thank you for the comment. This is also one of the reasons why we took the “point injections” performed with SOCOLv4 into account in this paper. The microphysical configuration and the timestep applied do not only matter for large injection rates but also for injection scenarios with smaller injection rates combined with more confined injection locations.

Line 235 pp: There is a mismatch of the names. You write very often CN and CN. One should be NC.

Thanks for pointing this out. We corrected this.

Fig 3: You average between 30N to 30S in Fig 2. Fig 3 shows a global average. To see the differences between the simulations you may add a 30N to 30S average in Fig 3.

The average between 30N to 30S at 50 hPa was only chosen because this represents the injection region. Therefore, averaging over this region best represents the aerosol formation processes without biases through advection and sedimentation of aerosols. However, for quantities such as effective radius, aerosol burden and radiative forcing it makes more sense to look at global averages.

Line 301 - 303: This is not true for Fig 4c. CN_20 is more similar to CN_200 compared to the burden plot. Why?

We changed to: “The latitudinal variations of the burden in Fig. 4a,b are reflected in the changes in radiative forcing (RF) in Fig. 4c,d, with reduced irradiance at high aerosol loading, and illustrate the direct radiative effects of the aerosol. However, in contrast to the smooth distributions of aerosol loading, RF exhibits a much higher degree of small fluctuations due to tropospheric cloud variability.“ The term “mirrored” might have been over exaggerated. However, CN_20 is only more similar to CN_200 over the south polar region. In a global average CN_20 clearly results in less RF compared to the other scenarios (see Figure 2). The largest contribution to the global average results from the tropics where CN_20 results in the smallest RF.

Line 312: Less time for ozone formation or stronger meridional transport? The last might be quite important.

You are right, it is rather the stronger meridional transport. We corrected this.

Line 321-322: Where? 6 to 24 DU are the values for the hemisphere, not at high latitudes.

Thanks for pointing to this. The values in the text were wrong. We corrected them.

Section 3.5: Pinatubo is not a good analogue for SAI. The injection rate is much higher as are the SO2 concentrations. It might be of interest for you to compare with Wrana et al (2023). After the eruption of Ulawun, satellite data show a decrease in particle size. So nucleation after the eruption is important to get a good agreement between model and data.

We do not claim that Pinatubo is an analogue to SAI. Volcanic eruptions are not a good analogue to SAI in general independent of the injection amount. This is due to the continuous supply of H2SO4 in SAI scenarios, which is not the case in volcanic eruptions (e.g. Heckendorn et al. or 2009 or Vattioni et al., 2019). We only have chosen Mt. Pinatubo eruption for sensitivity testing to the microphysical settings because it is a large eruption and thus, the effects of the microphysical settings become most apparent.

Line 399pp: Wan et al 2013 offer a solution of your problem: 'These errors can be significantly reduced by employing solvers that handle production, condensation and nucleation at the same time.' You should discuss this - employing in the model might be an even better solution. You may have a look for Wan et al (2023) as well (https://doi.org/10.48550/arXiv.2306.05377).

Indeed employing solvers which handle production, condensation and nucleation in parallel as described in Wan et al. (2013) would be optimal. We are thinking of implementing this into the next version of SOCOL. However, this would be beyond the scope of this project. We think the findings of Wan et al. 2013 are already discussed in enough detail in the paragraph you are pointing to.

Line 412-414: This is not true in general, only for the specific setup of the modes in combination with the injection strategy used in Laakso et al (2022).

Thank you, we added a side note: “In addition, the use of M7 with lognormal modes results in a minimum in the particle size distribution in the optimal size range for solar scattering due to the accumulation mode reaching its largest size, which adds mass to the coarse mode in the injection scenario applied in Laakso et al. (2022). The resulting gap between the two modes tends to underestimate gravitational settling.”

Line 415-416: spread: How is this related to this work. Aren't you comparing apples and pears here?

No, we do not compare apples with pears. We clearly state what we compare. It is important to highlight that different parameterizations of microphysical processes can result in substantial differences in resulting size distributions and thus, in resulting radiative forcing. Obviously, we can not directly compare the Lakso et al. 2022 study with our study due to different injection rates and injection scenarios. Therefore, we only point to the differences between models which apply different microphysical parameterizations. The differences in radiative forcing are even larger than the differences resulting from switching the calculation order and the radiative forcing in our model.

Line 424: As far as I understand Laakso (2022), SALSA does not use Vehkamäki. Nucleation in SALSA is much stronger than in HAM.

For SALSA Laakso et al. (2022) is pointing to Kakkola et al. (2018). However, there is no information about the type of parameterisation used in SALSA. In Kakkola et al. (2009)

state that SALSA uses the Vehkämäki scheme. Laakso et al. (2022) also points to Vignati et al. (2004) for M7, which states that M7 uses the Vehkämäki scheme.

Line 426: The collision rate is important for high SO2 concentrations. Otherwise the parameterizations of Vehkamäki (2002) might be not valid at all grid points. However, this is not well published. Määtänen et al (2018) is an upgrade and includes the collision rate. It might be worth to think about an implementation in your model.

Thank you for the comment. We will update the nucleation scheme to the one used in Määtänen et al. (2018) in the next version of SOCOL. See also answer to the following comment.

Line 443: Can you relate your results to Yu et al (2023)? You get very different answers with one nucleation parameterization, but different substepping etc. So, I wonder if this very general conclusion of Yu et al (2023) holds for your results. In Wrana et al (2023) we use Määtänen et al (2018). This parameterization reproduces the particle size after the Raikoke and Ulawun eruptions quite well. These small eruptions are much closer to SAI because they have a more similar eruption rate than the Pinatubo eruption. I recommend that you do a simulation, even if you do not want to include the results in this paper. You may gain more confidence, which is the better way to continue.

Yu et al. (2023) write: “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 the nucleation rate is high, a 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.”

It is unclear how our findings would affect the results reported by Yu et al. (2023). The solver in Yu et al. (2023) does not account for competition between nucleation and condensation and is semi-implicit together with a time step reduction for high H2SO4 supersaturations. The fact that nucleation consumes at most 1% of the sulfuric acid vapour is a good sign that the scheme is likely performing fine.

Indeed, the nucleation parameterization used in Määtänen et al. (2018) would likely be more appropriate to use in aerosol-chemistry climate models as it results in better agreement with observations. We will incorporate this in future versions of SOCOL. However, even though the local injection rate observed after smaller eruptions such as the Raikoke and Ulawun eruptions are more close to conditions of potential SAI scenarios, they are likely still not comparable to the aerosol size distributions resulting from continuous injections of SO2 in SAI scenarios (see Vattioni et al., 2019, or Heckendorn et al., 2009). However, we agree that we could learn from simulating these smaller eruptions and comparing them to observations and other models. We simply do not have the resources at the moment to do this analysis. We have added two sentences to the discussion section of the paper: “The reported weaknesses of the Vehkamäki scheme were addressed by Määttänen et al. (2018), who presented a new parameterisatio for sulfuric acid aerosol formation including homogeneous and ion-induced nucleation pathways validated by CLOUD laboratory measurements. The Määtäen et al. nucleation scheme which is reported to be valid for the whole range of atmospheric conditions including high stratospheric sulfuric acid concentrations during SO2 injection scenarios is recommended to be used in

aerosol-chemistry climate models instead of the Vehkamäki scheme (Määttänen et al., 2018).“

References

Määtänen, A., Merikanto, J., Henschel, H., Duplissy, J., Makkonen, R., Ortega, I. K., and Vehkamaki, H.: New Parameterizations for Neutral and Ion-Induced Sulfuric Acid-Water Particle Formation in Nucleation and Kinetic Regimes, J. Geophys. Res.-Atmos., 123, 1269–1296, https://doi.org/10.1002/2017JD027429, 2018.

Niemeier, U., Richter, J. H., and Tilmes, S.: Differing responses of the quasi-biennial oscillation to artificial SO2 injections in two global models, Atmos. Chem. Phys., 20, 8975–8987, doi.org/10.5194/acp-20-8975-2020, 2020.

Wrana, F., Niemeier, U., Thomason, L. W., Wallis, S., and von Savigny, C.: Stratospheric aerosol size reduction after volcanic eruptions, Atmos. Chem. Phys., 23, 9725–9743, https://doi.org/10.5194/acp-23-9725-2023, 2023.

References:

Walcek, C. J.: Minor flux adjustment near mixing ratio extremes for simplified yet highly accurate monotonic calculation of tracer advection, J. Geophys. Res., 105, 9335–9348, https://doi.org/10.1029/1999JD901142, 2000.

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.

Heckendorn, P., Weisenstein, D., Fueglistaler, S., Luo, B. P., Rozanov, E., Schraner, M., Thomason, L. W., and Peter, T.: The impact of geoengineering aerosols on stratospheric temperature and ozone, Environ. Res. Lett., 4, 045108, https://doi.org/10.1088/1748-9326/4/4/045108, 2009.

Vattioni, S., Weisenstein, D., Keith, D., Feinberg, A., Peter, T., and Stenke, A.: Exploring accumulation-mode H2SO4 versus SO2 630 stratospheric sulfate geoengineering in a sectional aerosol–chemistry–climate model, Atmos. Chem. Phys., 19, 4877–4897, https://doi.org/10.5194/acp-19-4877-2019, 2019.

Vignati, E., Wilson, J., and Stier, P.: M7: An efficient size resolved aerosol microphysics module for large scale aerosol transport, J. Geophys. Res.-Atmos., 109, D22202, https://doi.org/10.1029/2003JD004485, 2004.

Yu, F., Luo, G., Nair, A. A., Eastham, S., Williamson, C. J., Kupc, A., and Brock, C. A.: Particle number concentrations and size distributions in the stratosphere: implications of nucleation mechanisms and particle microphysics, Atmos. Chem. Phys., 23, 1863–1877, https://doi.org/10.5194/acp-23-1863-2023, 2023.