Dear Dr. Laakso,

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 manuscript authored by Vattioni, Stenke, et al. examines the impact of sequential operator splitting and the number/length of time steps on the simulated outcomes of climate intervention through stratospheric sulfur injections. Authors say in the manuscript that “The intention of this paper is to raise awareness within the (aerosol) modelling community for potential numerical problems within conventional aerosol microphysics modules” and it does that very well. The key finding and message for the modeling community is that, even when simulating the same scenario with an identical model and identical lines of code, results can vary significantly (in this instance, ranging from a radiative forcing of -2.3 Wm-2 to -5.3 Wm-2) due to the order in which the two processes are calculated. The aspect of this “choice” is a detail which is quite often overlooked in studies related to stratospheric aerosol injection and model intercomparison studies. Furthermore, the manuscript is exceptionally well-written, making it straightforward to read and devoid of any major issues. Consequently, I strongly recommend the publication of this manuscript.

I have only some minor comments or corrections. First more general ones:

I would like to see some discussion from the authors about the role of the coagulation in all of this. It clearly had an impact in Pinatubo simulations (which was a nice point by the way!), but does it have a major impact in the case of stratospheric aerosol injections? In these simulations coagulation was included in the microphysical subloop. What do you think, would it have had a major impact if the number of steps would have been increased only for nucleation and condensation, but not coagulation? And just to be sure: I do not expect to see any additional simulation or analysis on this, but it would be just interesting to know if the authors would have any thoughts about this. Usually coagulation is a computationally heavy process (however probably not that much in SOCOL, where the coagulation kernel is not calculated inside the subloop) and thus not wanted to be calculated more than is needed.

This is a very good comment. We looked at this early on and also performed some sensitivity simulations on this with the S25 simulations, but then we decided to not highlight this in the paper. In these sensitivity tests we looked at the role of nucleation/condensation vs. coagulation by applying a microphysical sub-sub loop in the S25 simulations. This means, in addition to the microphysical sub-loop, we applied another sub-sub-lop for either coagulation only (“5_coag” in Figure 1 below) or for nucleation and condensation only (“10_nuc_con” and “2_nuc_con” in Figure 1) within the microphysical sub loop. We performed two simulations with a microphysical sub-step of 20 - one with 5 additional sub-sub-step for coagulation (i.e. 20 x nuc/cond and 20x5=100x coagulation) and the other with 10 additional sub-sub-steps for nucleation and condensation (i.e. 20x10=200 times nuc/cond and 20 times coagulation).

As expected, the additional sub-loops for coagulation only (5_coag) does not significantly reduce the nucleation mode particles. This is due to the very large H2SO4 supersaturation resulting from only having 20 sub-loops for nucleation and condensation, which results in very large nucleation mode particle concentrations. However, in the accumulation mode the particle concentrations are clearly reduced, due to efficient coagulation.

Applying 10 additional microphysical sub-sub-steps for nucleation and condensation only (10_nuc_con), results in a total of 20x10=200 sub-steps for nucleation and condensation and only 20 for coagulation. Here, we observe a clear reduction of the nucleation mode particle concentrations, which is due to the lower supersaturation when having a smaller timestep for nucleation and condensation. However, due to the inefficient coagulation (20 sub-steps only) the concentration of the accumulation mode particles is too large compared to NC_200. This also explains why the nucleation mode concentration is even lower than in the case for NC_200: When we have large accumulation mode particle concentrations (immobile targets), coagulation with the tiny nucleation mode particles (very mobile projectiles) is more efficient.

To conclude, only increasing the timesteps of nucleation/condensation or coagulation does not result in satisfying results. Therefore, both, the timesteps of nucleation/condensation and coagulation must be reduced. In additional simulations we found that a microphysical timestep of 40 combined with an additional sub-loop for nucleation/condensation, resulting in 80 (i.e., 40 x 2) sub-steps for nucleation/condensation and 40 sub-steps for coagulation would be enough to be very close to the S25_NC_200 solution. As expected, nucleation/condensation required a smaller timestep since the timescale of coagulation is usually larger compared to the one of nucleation/condensation. Therefore, to save computer time, the number of sub-loops for coagulation need not be increased as much as the one for nucleation/condensation, but it should nevertheless be sufficiently small.

However, the “optimal” amount of “sub-steps” and “sub-sub-steps” is highly dependent on the scenario applied (spatial confinement of the injections, pulsed or continuous injections, injection location and time) and is for example very different for the Mt. Pinatubo case. Therefore, we decided to not show these results in the main manuscript, since this could result in the misleading conclusion, that there is an “universal optimal solution”.

We added the following sentence to line 234/235 of the new manuscript: “Since coagulation has the largest timescale and is computationally the most expensive process within the microphysical sub-loop, we also tested a scenario with 80 sub-steps for nucleation and condensation, but only 40 for coagulation (not shown).”

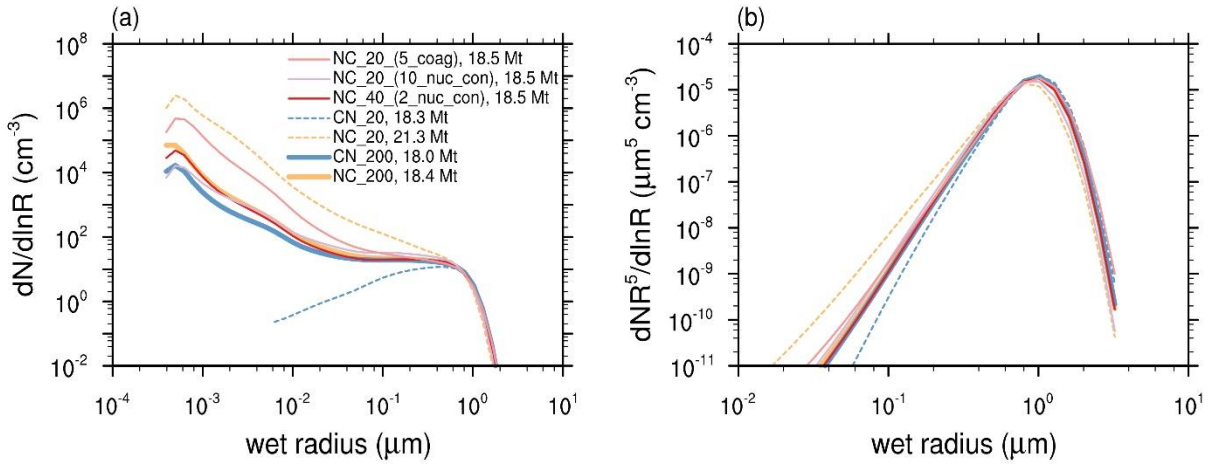

Figure 1: Sensitivity simulations of the S25 scenario for resulting size distribution (a) and “5th moment” (b) for an additional microphysical sub-sub-loop for nucleation/condensation only (nuc_con) and for coagulation only (coag). The burden in the legend indicates the total aerosol burden increase of each simulation compared to the reference run. The simulations shown were run with the same boundary conditions as the simulations in the main manuscript, with the only difference that here a one-year average of the year 2067 of a transient simulation is shown. This explains small deviations from the simulations shown in the main paper.

Authors also mentioned about the sensitivity of results to the chosen injection scenario, but this could be discussed little more in respect to take home messages of this study. What I mean is that e.g. increasing the number of timesteps, or calling nucleation routine first, had a large impact on S25 simulations but rather small for S5 simulations. Thus someone might think that this issue of calculating microphysics does not matter for 5 Tg(S)/yr injections. However, in this study the injections were done in a quite wide area/band (30 N - 30 S latitudes) and I assume there would have been a larger difference if the injections would have been done to a narrower band. This was actually shown more or less by ESM SOCOLc4 simulations for S5p.

Thank you for pointing this out. The conclusions could indeed have given the wrong impression that this issue only applies to large injection rates, which is wong as we show with the S5p simulations. We included the “S5p” results with SOCOLv4 in the last bullet point of the conclusions. And also, we put more focus to this aspect in the second last paragraph of the conclusions now:

“It should be emphasized that our conclusions are mainly based on simulations of "regional" SO_2 injections, which are supported by "point injection" scenarios and simulations of the 1991 Mt. Pinatubo eruption. As the nucleation rate strongly depends on the gas-phase H_2SO_4 concentration, ambient temperatures and relative humidities, the optimal number of microphysical (sub-)timesteps will depend on the assumed SO_2 injection rates, but also on the injection scenario and spatial confinement of the injections. "Point" injections of SO_2, for example result in very high, but locally confined H_2SO_4 supersaturations, which makes the results more sensitive to the details of the microphysical approach. This effect is shown with the "point injection" scenarios (S5p), which are much more sensitive to the microphysical settings compared to the "regional injection" scenarios (S5, see Fig. 3b).”

Some specific comments:

Some lines in the text there are “CN” while it was clearly referring to “nucleation first”/NC. Please check these (or neglect me if I have understood something wrong):

P9 L209 "nucleation first" (CN) -> "nucleation first" (NC)

P9 L214 “of the CN and CN simulations” -> “of the CN and NC simulations”

P10 L239 “CN and CN” -> “CN and NC”

P10 L245 “(CN, “ -> “(NC, “

P11 L251 “CN and CN” ->”CN and NC”

P12 L253 “CN_20 setting” ->”NC_20 setting”

P12 L265 “sequence (CN_20)” ->

P12 L282, P13 L294,

P14 L335, L247,

P16 L355, L361, 363-365

Fig5 text

P18 L416

Thanks for pointing to the NC and CN chaos. We corrected all wrong abbreviations and updated the manuscript accordingly.

I noticed that this is something that Daniele (RC1) commented on already but I will mention it anyway: In the introduction it is said that “climate intervention” is used instead of “geoengineering”, but still “geoengineering” is used in a couple of lines in the text.

Thank you for pointing to this. We made the terms consistent and use “climate intervention”

P1 L15 “25 MT/yr” -> “25 Tg(S)/yr” and “timesetp” -> “timestep”

We corrected this and made it consistent.

P5 L137 “H_2O_2” -> “H_2O”?

We corrected the typo to “H_2SO_4”.

P6 L159 Maybe you could add that one major difference between SOCOL model versions is also the atmospheric model (ECHAM5 / ECHAM6). If I am not totally wrong.

ECHAM6 is part of MPI-ESM1.2, but we included a note that SOCOLv4 is based on ECHAM6 to make it clear.

P7 L177-184 I have to admit that I don't always completely remember all GeoMIP scenarios but here it is said that the simulated point scenarios are following the G4 GeoMIP scenario of Kravitz 2011. However G4 is based on RCP4.5 and 5 Tg SO2/yr is injected (= 2.5 Tg(S)/yr). I assume you meant to refer to some other GeoMIP experiment? I was also thinking that “point” might be slightly misleading as the injections are done along several grid points along the meridian and thus it is different from in Pinatubo simulation. But there is no perfect way to name these and I do not have a better suggestion for the name.

We specified as follows:

These “point emission” scenarios followed the G4 GeoMIP scenario described in Kravitz et al. (2011). However, instead of RCP4.5 GHG and injections of 5 Mt SO_2 /yr as specified in Kravitz et al. (2011), we used SSP5-8.5 GHG and injected 5 Mt/yr S in the form of SO_2, which is consistent with Wunderlin et al. (2024). The transient SSP5-8.5 boundary conditions allow us to explore the sensitivity of surface temperature to the call sequence in a fully coupled ESM.

P 7 L 190-193 You could describe the Pinatubo experiment also here in text as it is done in Table 1. I mean how much SO2 is injected, which altitude and which ISAMIP experiment you are referring to. You could also mention why you chose “low-shallow-injection scenario”.

Thanks for pointing to this. We specified:

“In the absence of observational data of the stratospheric aerosol layer under climate intervention conditions, we also tested the effect of different microphysical settings in the modeling of the 1991 Mt. Pinatubo eruption. The 1991 Mt. Pinatubo eruption was specified

as 5 Tg S emitted in the form of SO_2 at 21-23 km altitude (2 model levels) above the Mt. Pinatubo geographical location (i.e., two model grid boxes) during one day. This set up corresponds to the HErSEA_Pin_El_Ism scenario proposed by the Interactive Stratospheric Aerosol Model Intercomparision Project (ISA-MIP, Timmreck et al., 2018), which has been shown to have better agreement with observations for some variables in (Quaglia et al., 2023) compared to scenarios with larger emission amounts and different emission altitudes"

P10, Fig. 2 Please check that (a)-(f) in the text (description of the figure) corresponds to the ones in the figure.

Thanks for spotting, I corrected this.

P11, Fig. 11. I recommend using some other color than light blue for optimal effective radii or at least make it darker. I did not see it when I printed the manuscript and it is not very clear in the pdf.

We changed the color to light green and made it less transparent.

P11, Fig. 11 or P12 L260, This probably would not need new figures and can be just mentioned in the text but it would be really interesting to see individual radiative forcing for shortwave and longwave radiation separately. I would be expecting that the difference in LW radiative forcing between simulations is relatively small. By the way, if radiative forcing is calculated as difference of radiative fluxes between perturbed and control/background scenarios, and not by e.g. double radiation call with and without aerosols, you might see some change in LW radiative forcing due to the land temperature adjustment which might be relatively large in case of 25 Tg(S)/yr injections. Of course this is something that does not affect your conclusions, but good to consider.

Yes, you are right (see Figure 2 below). The LW RF is very similar between simulations. There is also no significant difference between the S5 and the S25 simulations (see Figure 2 below) and thus, no significant effect from the land adjustment in the LW.

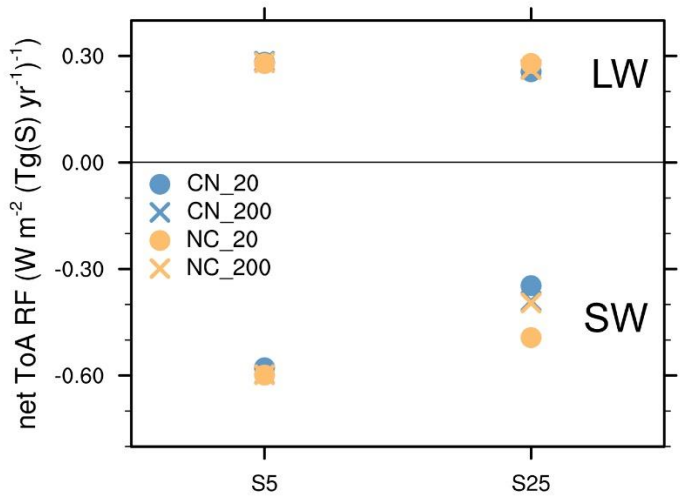

Figure 2: This Figure shows the SW and LW RF efficiency (RF/Mt injected material). The LW efficiency are the upper points and crosses (positive numbers) and the SW efficiency are the lower points and crosses (negative numbers).

P12 L284 This is more just a comment, but it is really interesting that the difference in temperature response between CN and NC scenarios is quite large here. As I mentioned

above, I am expecting the difference to be caused by the fact that the latitude band is narrower than for SOCOL-AERv2 simulations. In Laakso et al. 2022 point injection led to more similar response between SALSA (prefers nucleation) and M7 (prefers condensation), but there sulfur was injected to the one model grid point which probably gave nucleation more suitable conditions to fight against condensation in M7 simulations.

Yes, this is also our suspicion. The “point” injections (S5p) result in about 1000 times more confined injection regions (10°N to 10°S at one latitudinal band instead of 30°S to 30°N at all longitudes) and thus larger local H_2SO_4 concentrations. Nucleation increases exponentially to larger H_2SO_4 supersaturations and is thus much stronger in regionally more confined injection scenarios. As mentioned in your second major commend above, we have now highlighted this artefact more in the revised manuscript. However, it should also be kept in mind that also the model resolution is different between the models, which can also result in different results.

P13 L317 I am not sure if I understood how nudging of winds was done. I assume it was not fully nudged if there are changes in atmospheric circulation?

SOCOL-AER simulates the wind fields interactively. However, the model version with 39 vertical pressure levels does not generate a QBO. Therefore, the QBO is artificially imposed by a linear relaxation of the simulated zonal winds in the equatorial stratosphere to observed wind profiles over Singapore perpetually repeating the years 1999 and 2000 (due to a time slice set up in our main case). Thus, SOCOL-AER simulates its own wind fields, but the QBO is nudged towards observed wind fields. See Stenke et al., 2013 for details.

P18 L414. Actually, in M7 simulations of Laakso et al. 2022 growth of the particles in accumulation mode was not restricted, but the size of the accumulation mode was. I mean that accumulation mode was in its maximum size, and when particles in accumulation mode grew up (by condensation or coagulation), they were transferred to coarse mode. This created a gap between these two modes.

Thanks for pointing this out. I changed to “In addition, the use of 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. The resulting gap between the two modes tends to underestimate gravitational settling.”

References:

Kravitz, B., Robock, A., Boucher, O., Schmidt, H., Taylor, K. E., Stenchikov, G., and Schulz, M.: The Geoengineering Model Intercomparison Project (GeoMIP), Atmospheric Science Letters, 12, 162–167, https://doi.org/10.1002/asl.316, 2011

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Wunderlin, E., Chiodo, G., Sukhodolov, T., Vattioni, S., Visioni, D., and Tilmes, S.: Side Effects of Sulfur-Based Geoengineering Due To Absorptivity of Sulfate Aerosols, Geophysical Research Letters, 51, e2023GL107 285, https://doi.org/10.1029/2023GL107285, 2024