Review of "*Exploring accumulation-mode-H2SO4 versus SO2 stratospheric sulfate geoengineering in a sectional aerosol-chemistry-climate model*" by S. Vattioni et al.

Comments by anonymous reviewer #2 are in bold. Author responses are in blue.

Manuscript by Vattioni et al studies several injection strategies for stratospheric sulfur geoengineering with gas phase SO2 and sulfate droplet injections. Research is done by using global 3D-aerosol-chemistry-climate model. This is one of the few studies where the impacts of stratospheric sulfur injections are studied with a sectional aerosol model coupled with/included in a global climate model. Authors have simulated several different scenarios to cover a wide range of options to inject sulfur to stratosphere.

Even though the general idea of the study and the studied scenarios are not totally new, the study shows several eye-opening results and it is a good addition to existing research. Currently there are relatively few studies where stratospheric sulfur geoengineering is simulated by including aerosol microphysics and especially with sectional aerosol model. Radiative forcing of stratospheric sulfur geoengineering is dependent on several factors, related to how sulfur is injected, but also how the microphysics is modelled. Thus, it is valuable to get information from different scenarios simulated with different models. Authors also quantify microphysical processes (such as nucleation, condensation, coagulation) in various scenarios which helps to understand the impacts of microphysical processes on geoengineering. In addition, for example, the responses in OH concentration were surprising, but well justified. Overall this is an interesting and excellent study. It is well written and does not leave open questions. Thus, I recommend publishing this manuscript and I have only minor comments on some specific points in the text. I also have to say that it is quite impressive that the work is based on a master's thesis.

I want to mention that I do not agree with reviewer 1 concern about differences between longitudinal distribution of emitted sulfur. As it is generally known, and pointed out in this study, results from point like simulations do not differ much from injections over all longitudes. In addition, it would be challenging to do an apple to apple comparison between the results of this and earlier studies, and I think it is not necessary in this case. My opinion is that the author's choice to use "all longitudes" -case as "default" option and pulsed scenario as a sensitivity case would have been natural choice for me too.

We would like to thank reviewer #2 for her/his insightful comments and suggestions. Please find our detailed replies below:

P1, L17: As was already commented by Alan Robock, using parentheses like this is a bit confusing.

Corrected.

P1, L22-23: "Increasing the local SO2 flux in the injection region by pulse or point emissions reduces the. . ." Would it be better to say something like: "concentrating injections to smaller regions by pulse or. . ." You don't just increase emissions somewhere but simultaneously decrease (remove) them elsewhere.

Was changed to: "In the case of SO_2 emissions, limiting the sulfur injections spatially and temporally in form of point and pulsed emissions reduces the total global annual nucleation… "

P2, L20: I would include following citation: Niemeier, U. and Timmreck, C.: What is the limit of climate engineering by stratospheric injection of SO2?, Atmos. Chem. Phys., 15, 9129-9141, https://doi.org/10.5194/acp-15-9129-2015, 2015. It shows nicely the reduced efficiency in the case of really high loading.

The citation was added.

P2, L22: Is it (i.e. point 4) **really a limitation? This study is not concentrating on this topic so this sentence can be removed.**

Our original "point (4)" has been removed as it is not a limitation in terms of radiative forcing.

P4, L2 Just a comment, sigma is usually fixed and same mode width does not represent well both coarse mode particles in troposphere and stratosphere (long living particles).

Corrected.

P4, L8: Sectional aerosol model is also used in:
Laakso, A., Kokkola, H., Partanen, A.- I., Niemeier, U., Timmreck, C., Lehtinen, K. E. J., Hakkarainen, H., and Korhonen, H.: Radiative and climate impacts of a large volcanic eruption during stratospheric sulfur geoengineering, Atmos. Chem. Phys., 16, 305-323, https://doi.org/10.5194/acp-16- 305-2016, 2016.
Laakso, A., Korhonen, H., Romakkaniemi, S., and Kokkola, H.: Radiative and climate effects of stratospheric sulfur geoengineering using seasonally varying injection areas, Atmos. Chem. Phys., 17, 6957-6974, https://doi.org/10.5194/acp-17-6957-2017, 2017.

The references to the two paper have been added, including a short discussion about their model.

P4, L18: "the radiation scheme did not interact with the aerosol module" This is not true.

This part of the sentence was removed.

P6, L11: Is there some explanation behind the decision to use 1.83 MT S yr-1 injections? For me it sounds like an accidental choice where you originally planned to do injections with certain mass but after all simulations were done, you noticed that unit in emission(/injection) was not what it should have been. However, I do not say that this is a problem, because there is not anything "wrong" to use this value, but if there is a sensible reason for use this specific value, it should be mentioned. This is also just a comment, but it would have been nice to see differences between SO2 and sulfate injections in a case of larger amount of injection.

The initial idea was to emit 2 Mt/yr, but due to a problem in the emission scheme, effectively only 1.83 Mt/yr have been emitted. As you already mentioned, this has no effect on the conclusions made in this paper as the same amount of sulfur has been emitted in all our model simulations. The 1.83 Mt/yr correspond to 9-20% of the emitted amount of sulfur from the Mt. Pinatubo eruption, depending which model one considers as the estimate of total S emission ranges from about 10-20 Mt S. Unfortunately, we did not have the resources to conduct simulations with different emission magnitudes. We clearly declare that the sensitivities with respect to aerosol burden and radiative forcing vary for different emission magnitudes.

P6, L13: There are several estimations for mass of the emitted sulfur from Mt Pinatubo eruption. It would be good to cite some study.

We used Sukhodolov et al. (2018) as a reference, who conducted a Pinatubo study with SOCOL-AER.

P6, L21: QBO nudging (without nudging winds generally) is new to me. If you can open this method by few clear sentences, it would be great. If not, then it is ok as it is.

I changed the sentence to: "The QBO was taken into account 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."

P8, L28 and L33: Based on table 2, I got 26.8% shorter resid. time in GEO_SO2_15 than in corresponding AERO-case (not 23.3%). What is 32% difference in L30? It would not be the first time that I cannot calculate something right but please check these.

You are right. Thank you very much for spotting this mistake. All calculations in the paper have been recalculated and corrected if necessary.

P9, L2: and maybe due to the coagulation?

Corrected to: "In the AM-H_2SO_4 case, the number concentration of nucleation mode particles decreases below background conditions due to the increased surface area available for condensation (Fig. 2) and increased coagulation of nucleation mode particles with accumulation mode particles."

P9: It would be useful if radiative forcing for LW was mentioned at some point. Kleinschmitt et al. 2018 got quite large LW forcing values compared to other studies and it would be interesting to see how this is in the model used in this study. I expect that there is not much (absolute) difference between cases where sulfur is injected as SO2 or sulfate (?).

We did not look at LW radiation as we used constant sea surface temperatures. We only observed surface LW radiation anomalies of <0.1 W/m^2. For a solid LW anomaly estimate a coupled ocean module would be necessary.

P10, L4: Reduction is seen only in clear-sky forcing but not in all-sky.

Corrected.

P10, L6: As was pointed out by reviewer 1 too, I had to google "surf zone" so maybe it is not that familiar word.

Was corrected to "…emitting partly into the stratospheric surf zone and not only into the tropical pipe. The stratospheric surf zone is the region outside the subtropical transport barrier where breaking of planetary waves leads to quasi horizontal mixing (McIntyre and Palmer, 1984; Polvani et al., 1995)."

P10, L21: It is better to use 25km instead of 24hPa to be consistent with experiment names.

Changed.

P11, L2: 0.95 -> 0.095 um

Corrected.

P11, L11: compared to . . .

Compared to the pure AM-H2SO4 emission scenario (i.e. GEO_AERO_15). Corrected.

P12, L5: Based on my experience, aerosols are not affecting much on LW fluxes at the surface. This line ("The longwave surface. . .") can be removed.

Removed.

P12, L14: "constant climatological SO2"? What does it mean?

Corrected to: "Kleinschmitt et al. (2018) applied a mean lifetime of 41 days for SO_2 to H_2SO_4 conversation in their study and found a SO_2-to-H_2SO_4 conversion rate of 96 %."

P12, L21: Parenthesis thing - same as in abstract

Corrected

P12, L29-30: Just a comment: I don't know has this been pointed out in some earlier studies, but if it has, at least I have missed it. This was an interesting remark and it sounds credible. In addition, the size distribution of particles is different in tropical peak compared to higher latitudes.

Thank you for the comment. I did not find other studies which made this point in context of solar geoengineering. I added a sentence pointing to the different size distributions in tropical and higher latitudes.

P13. L18: "...the smaller the negative side effects" Can you really say this? There are several negative side effects which are not studied here.

I clarified this by adding: "…the smaller the investigated negative side effects…"

P14, L29: "are only increased by about 4%" I would remove word "only". I was surprised that OH concentration was generally increased.

Corrected

Figures: In addition to reviewer 1 comments please correct following typos:
Fig3: Areosol -> aerosol (in upper right)

Corrected

Fig6: Janauary -> January

Corrected