Response to Reviewer 1

We thank the reviewer for careful reading of our manuscript and constructive comments. Our responses to specific minor comments are given below in italics following each comment. Considering the issues raised by the reviewer has allowed us to improve our manuscript by clarifying these issues in the text. We are grateful for the reviewer's time and thoughtfulness.

Review for Weisenstein et al.,

"A model intercomparison of stratospheric solar geoengineering by accumulation mode sulphate aerosol"

Submitted ACPD

Here authors analyse CCM output from a dedicated GOIP solar engineering experiment "AM-H2SO4". This experiments is designed to inject geoengineering Sulphur (S) in the stratosphere in terms of particles (SO3 or H2SO4), so that stratospheric aerosol particles would grow mainly in accumulation mode, thereby negating effects of faster particle growth (and associated particle sedimentation). Analysis in this manuscript suggest that only three CCMs (WACCM, ECHAM5-HAM and SOCOL-AER) managed to complete these simulations. Basic idea behind these simulations is to differentiate model response to the SO2 vs particle injection under different (5 vs 25) Tg S injection magnitude scenarios. Authors find that all three models show increased radiative efficacy (in terms of radiative forcing) when Sulphur is injected in "AM-H2SO4" mode compared to gas phase injection. Also sensitivity simulations with different injection patterns (two points at 30° N and 30° S vs injection in a belt along the equator between 30° S and 30° N) find opposite response.

Overall this is well written manuscripts and fits well within ACP scope. Hence, I will like to recommend this manuscript for the publication with minor corrections.

Minor Comments:

1. Page 3: Line 28: Does that mean ECHAM has identical ozone loss in all the simulations?

   Echam uses the same prescribed ozone and OH fields for the reference and geoengineering simulations. Thus there is no ozone loss due to geoengineering in the simulations. We have made this explicit in the text.

2. Line 6 Line 18: I am really surprised that you use only 2 year spin up period. If you plot global burden, you would see steady increase in burden before curve

flattens, depending on dry and wet deposition schemes. Unless you have meteoric smoke particles transporting or mopping S-containing species downwards and there is lack of particle evaporation (temperature increase due to ozone increase), gas phase tracers (e.g. SO2, H2SO4) would show steady transport upwards. Overall tracers should reach to equilibrium state near model top after 3 to 4 years as they transport downward in the polar vortex. I think that is why WACCM (page 10 line 8) shows increasing residence with increase in injection amount. For e.g. Dhomse et al., 2013 (Figure 3) equilibrium for meteoric smoke particles is about 10 years. I suspect it should be at least 5 years for these simulations.

While we expect that aerosol concentrations near the top of the sulfate layer and at high latitudes might still be evolving after 2 years, this paper analyzes only globally averaged quantities which we find to be fairly stable with time after 2 years for most scenarios. See enclosed plots of time evolution of global burden for the CESM2 and MAECHAM5-HAM models. Aerosol global burdens are still rising after 4 years with AM-H2SO4 injections of 25 Tg/yr but are stable after 2 years for the other scenarios. The SOCOL model simulations actually used a 5 year snmi-up period and then the following 8 years are averaged, so those results should not have an issue with spin-up length. For the CESM2 and MAECHAM5-HAM models, global aerosol burdens using averages of the last 5 or 6 years of the 10 year simulations are greater than for the 8 year averages by only 2-3% with AM-H2SO4 injections of 25 Tg/yr. We will acknowledge in the paper that the spin-up period was too short for these scenarios but has minimal effect on any of the quantities presented or conclusions drawn.

None of these simulations contains meteoritic smoke particles. The residence times of aerosol shown in Figure 3 of the paper are derived from burdens and injection rates (not diagnosed removal rates), with injection rates constant in time. Thus correcting for a too-short spin-up time in the AM-H2SO4 25 Tg/yr scenarios would increase the residence time for the 25 Tg(S)/yr injections in our plot and does not explain why CESM2/WACCM shows increasing residence with an increase in injection amount.

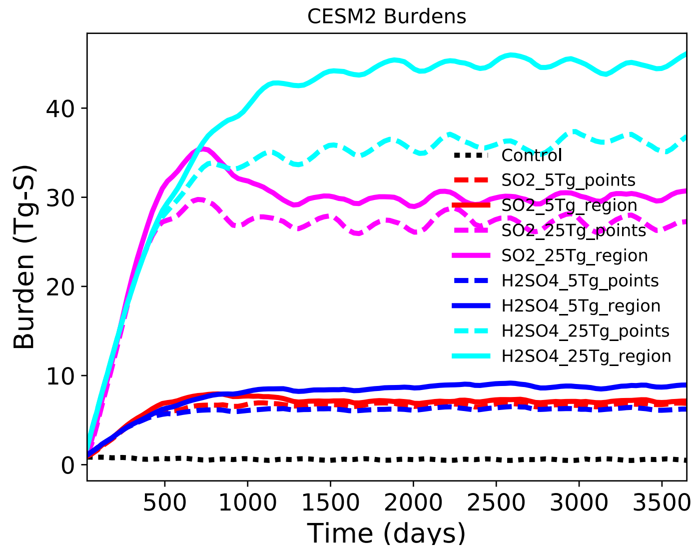

Figure R1: Time evolution of global aerosol burden in the CESM2 simulations.

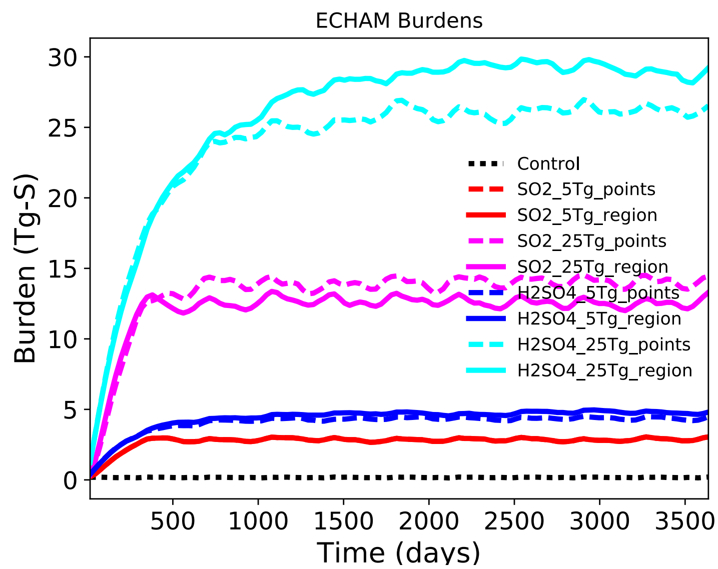

Figure R2: Time evolution of global aerosol burden in the MAECHAM5-HAM simulations.

3. Page 6: Line 19: What is baseline or reference simulation? Do you mean from respective SSP8.5 simulation? Is it from a single ensemble member or from ensemble mean?

The baseline scenario is described in the preceding paragraphs with SSP8.5 2040 GHGs and ODSs and SSTs for the 1988-2007 period. It is from a single ensemble member and we use an 8-year average from each model in our analysis.

4. Page 8 : line 1: Are you sure about only 10%? One needs to have very fast wet deposition. I think you should provide a line plot showing time variation in global burden.

   The tropospheric burden is found to be 10% or less of the global aerosol burden increase in the SOCOL-AER model, even for the 25 Tg(S)/yr cases. Tropospheric burdens are not available for the other models.

   A line plot of global burden time variation is provided for the reviewre (figures R1 and R2 herer, but as we don't have saved data of the 5-year spin-up period for SOCOL-AER, we do not include these plots in the paper.

5. Page 9: line 1: it should be other way round : weaker stratosphere troposphere exchange in the SH hence more aerosol accumulate in SH mid-lats.

   This referred to the lack of mixing between mid latitude and polar air in the southern hemisphere and hence the aerosol burden contrast in those two regions, which is not seen in the northern hemisphere with its weaker polar vortex and more efficient mixing to the pole. We modified the text to make this explicit.

6. Page 11: Figure 4: Does slope remain constant if you use only last 5 year data (5 year spin up).

   Yes, using the last 5 years produces minimal impact of the figure.

7. Page 12: line 9 : Any idea why ECHAM shows much weaker sensitivity.

   We are not able to diagnose the precise reason or reasons for the weaker sensitivity in the ECHAM model. Previous comparisons between ECHAM and CESM pointed to weaker tropical upwelling in ECHAM than CESM. The models also differ in the details of their aerosol formulations and their chemistry.

8. Page 18 : line 6 : Edit : 30°S-30°N

   We have corrected this.

9. Page 21: line 19: Are you sure it is minor. In Dhomse et al (2015), it is about 3%. With significant Cly decrease, future ozone losses would be largely controlled by NOy chemistry (e.g. Ravishankara et al.,2009), I would expect up to 5% ozone increase in the tropical middle stratosphere.

We do discuss ozone control by NOy chemistry and have added the reference to Ravishankara et al. (2009) and additional discussion regarding future ClOx and NOx changes. Ozone increases due to SO2 and AM-H2SO4 injections in these models are found in the 10-50 hPa region and near the tropical tropopause in the CESM model.

References:

Dhomse, S.S., Saunders, R.W., Tian, W., Chipperfield, M.P. and Plane, J.M.C., 2013. Plutoniumâ⓪❺238 observations as a test of modeled transport and surface deposition of meteoric smoke particles. Geophysical Research Letters, 40(16), pp.4454-4458.

Dhomse, S. S., M. P. Chipperfield, W. Feng, R. Hossaini, G. W. Mann, and M. L. Santee (2015), Revisiting the hemispheric asymmetry in midlatitude ozone changes following the Mount Pinatubo eruption: A 3-D model study, Geophys. Res. Lett., 42, 3038–3047, doi:10.1002/ 2015GL063052.

Ravishankara, A.R., Daniel, J.S. and Portmann, R.W., 2009. Nitrous oxide (N2O): the dominant ozone-depleting substance emitted in the 21st century. Science, 326(5949), pp.123-125.

Response to Reviewer 2

We thank the reviewer for careful reading of our manuscript and constructive comments. Our responses to general and specific comments are given below in italics and indented following each comment. Considering the issues raised by the reviewer has allowed us to improve our manuscript with more precise wording. We are grateful for the reviewer's time and thoughtfulness.

Review of manuscript "A Model Intercomparison of Stratospheric Solar Geoengineering by Accumulation-Mode Sulfate Aerosols" by Debra Weisenstein et al.

This manuscript presents results from a model intercomparison comparing interactive stratospheric aerosol simulations within co-ordinated multi-model experiments to explore the global dispersion and radiative forcing that would result from a continuous source of sulphur dioxide or accumulation mode sulphate aerosol particles with two different emissions scenarios: none emitting only at 30N and 30S, the other as a constant source between 30S and 30N.

The intercomparison compares results from 3 different interactive stratospheric aerosol models (WACCM-MAM3, MAECHAM5-HAM and SOCOL-AER), and represents a potentially very interesting contribution to understand the predictions from the models, each having differing sophistication in their aerosol modules, and in the vertical and horizontal resolution of the GCM's advection.

Whilst the results are interesting, and certainly will be publishable in a revised form, the aim and design of the model experiments are surprisingly poorly described, and the Introduction and interpretation need to include some discussion also of the tropical stratospheric reservoir, in relation to the differences between the two scenarios.

In several places the manuscript has unscientific language and vague statements that need to be changed to terms more appropriate to a journal article. For example "may produce overly large aerosols" (page 1, line 12) and "unfavorable aerosol size distributions" (page 2, line 20) and "Our aerosol size distribution" (page 3, line 14) are clearly subjective terms that need to be better phrased to communicate the issues involved.

> *We have modified the writing to be more scientifically precise and thank the reviewer for pointing this out.*

There are also a few places where the wording is poor, for example "These limits might be addressed" (page 2, line 25), "some of these limits may be addressed by altering the size distribution of sulfate aerosol" (page 2, lines 26-27). The authors are clearly aware that these issues are at the heart of the science to understand the efficacy and risk of a hypothesised large-scale injection of precursor gas of idealised particle for solar radiation management. Referring to "altering the size distribution of the sulfate aerosol" grossly simplifies the complex interplay of processes involved -- and the wording needs to communicate consistently with an awareness of these issues.

I also find it very surprising that, in this initial version of the manuscript, the authors have not adequately explained the rationale for the very interesting model experiments they are presenting results from. The two "injection scenarios" presented in the paper: 1) emitting continuously at two sites at 30N and 30S, and 2) emitting continuously throughout all latitudes between 30N and 30S, not surprisingly cause very different enhancements to the stratospheric aerosol layer (as is clearly seen in Figure 2). The 30N and 30S two-site scenario causes the stratospheric aerosol layer enhancement to be almost exclusively in mid- and high-latitudes, with only a very minor elevation in stratospheric aerosol optical depth in the tropics. The main reason for this is of course that the two-site injection scenario emits SO2/particles entirely outside the tropical stratospheric reservoir (between 20S and 20N). It is well established (e.g. Dyer, 1974) that the residence time for volcanic aerosol clouds formed in the tropical stratosphere are much longer (around 2 years) than for eruptions forming stratospheric aerosol clouds in the mid-latitudes. The reason is the continuing tropical upwelling and the transport barrier at the edge of the tropical pipe, and analysis of satellite measurements in 1991-1992 show the effect for example on the dispersion of the Pinatubo aerosol cloud (see Grant et al., 1996 for example). There needs to at least be sentence briefly mentioning the tropical stratospheric reservoir in the Introduction, and some discussion of the seasonal cycle of the Brewer-Dobson circulation (e.g. as set out originally by Dyer et al., 1968 for the Agung aerosol cloud).

Just to be clear, my review is not saying these results are not interesting, the results are indeed very interesting --- and this is a laudible effort to have a set of experiments to better understand any differences in predictions with the models -- but there needs to be a much clearer explanation of the rationale for why these scenarios were chosen.

It's implicit in the text that the 30N and 30S case might represent a limited 2-site injection strategy, but it needs to be made clear that the two scenarios are not really comparable, and that our understanding of stratospheric circulation would clearly mean that the 2-site 30N and 30S injection scenario would give a mid-latitude focussed stratospheric aerosol forcing, whereas the 30S-to-30N area-source scenario is presumably designed to give a more evenly-spread stratospheric aerosol layer enhancement, with then substantial radiative forcing also in the tropics.

> *We have modified the manuscript to more fully describe the rationale for the regional and 2point injections in both the abstract and scenario descriptions.*

I'm also recommending the authors consider changing the title, because the term "Solar Geoengineering by Accumulation-Mode Sulfate Aerosols" is not consistent with what the authors state that particular model experiment is representing. Firstly, the models ran separate simulations with continuous emission of SO2, in addition to the experiment with the particle source, so the experiments are to explore also the injection of SO2, in addition to direct injection of particles. So the title should either state that both are carried out, or else just give a more general summary-term there.

> *We have modified the title to read: "An aerosol-climate model intercomparison of stratospheric solar geoengineering by injection of SO2 or accumulation-mode sulfate aerosols".*

Secondly, the manuscript states (page 3, lines 14-17) that the particle-injection experiment is designed to represent particle sizes that would occur at the grid-scale of the large scale models following injection of SO3 or H2SO4 from high altitude aircraft, a localised plume subsequently generating a source of accumulation mode particles at the grid-scale of the GCMs. The text states "Our aerosol size distribution is consistent with Pierce et al. (2010) and Benduhn et al. (2016) who modelled plume microphysics and found that injection rate could be adjusted to produce sulfate aerosol size distrihbutions in the 0.1-0.15 micron radius size range."

I'd say first that I'm not sure either of those two first authors would argue that one can simply adjust the injection rate to produce the desired size distribution. I would expect that both would explain that there would be a substantial variability in the size distribution generated as the plume subsequently entrains into, and becomes mixed with the surrouding ambient air. So I'd argue that the text "could be adjusted to produce" is not really adequately representing the eventual variability in sizes that would result there. That said, I accept that a large diversity range for particle size is given (0.1 to 0.15 microns). It's again a case of the wording not adequately communicating the issues involved.

> *We have modified the wording here: "Our aerosol size distribution is consistent with Pierce et al. (2010) and Benduhn et al. (2016) who modelled plume microphysics and found that sulfate aerosol size distributions in the 0.1-0.15 μm radius size range could potentially be produced. Detailed modelling of this complex process for a full range of stratospheric physical, chemical, and microphysical conditions awaits further studies."*

In my specific comments below, I'm recommending the authors consider using the terminology "sub-grid-scale sulphate emission" rather than "accumulation-mode particle emission", or as an alterntive they could actually explain the rationale of the experiment is to represent a proxy for an idealised particle source, deliberately designed to produce particles at a particular desired size.

In light of the likely large variability in particle sizes that continued gaseous emission of SO3 or H2SO4 would cause, to me it is actually this engineered particle-emission scenario that these controlled size-distribution experiments are representing.

We now acknowledge this: "These GCMs can effectively simulate changes in global aerosol burden, radiative forcing, ozone, and stratospheric temperature and circulation. As input, they would take the particle size distributions from aircraft plume model studies but could represent any hypothetical input of particles. The input size distribution is simplified here by using a single mode radius and mode width at all grid points and times."

The other similar terminology issue I identify in my specific comments, is that the authors seem to use both the acronym "SRM" for solar radiation management, and also use the acronym "SSG" for stratospheric solar geoengineering. In my view, the paper needs to be consistent in either using SRM or SSG, but not both. My recommendation would be to use the acronym SRM, since the acronym SSG is often used for "scientific steering group", and SRM is also (in my mind) the more established term.

We have replaced "SSG" with "SRM" throughout the paper for consistency.

I'm suggesting the title should also be clear these are interactive stratospheric aerosol simulations being intercompared, with my suggestion being to change the title from "A model intercomparison of stratospheric solar geoengineering by accumulation-mode sulfate aerosols" instead to something like: "A co-ordinated intercomparison of interactive stratospheric aerosol model experiments for hypothesised scenarios of solar radiation management by sulfate aerosols"

We have modified the title to read: "An aerosol-climate model intercomparison of stratospheric solar geoengineering by injection of SO2 or accumulation-mode sulfate aerosols".

I provide below a list of specific comments I am asking the authors to address, and with these comments requesting a change in the tone of the narrative of the manuscript, my review then finds major revisions are needed. The authors may find it relatively easy however to make these changes -- with the Figures, and much of the results section is in good shape, requiring only minor revisions.

Specific comments:

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1) Page 1 -- lines 1-2 -- Further to the comments above, I strongly recommend the authors consider using a different term than "solar geoengineering by accumulation-mode sulfate aerosols".

The optical depth from the stratospheric aerosol layer mainly comes from sulfate aerosols in the accumulation mode size range, and the forcing from any geoengineered enhancement to the stratospheric aerosol layer would be caused by particles in the accumulation mode part of the size spectrum. So using the precursor term "accumulation-mode" prior to the "sulfate aerosols" is not really a useful descriptor of the effect.

I realise that one of the co-ordinated multi-model experiments involves each model adding a continued source of sulfate aerosol particles at a particular constant size distribution (in the accumulation mode size range) but that term is then referring to some specifics of the design of the model experiment.

Remember that the residence time of particles in the stratosphere is months to years and the resulting size distribution from a continued emissions is rather a response to that source of particles, and the microphysical and dynamical processes likely mean the resulting size distribution may differ substantially from that within a localized primary emission. That does not necessarily rule out devising a source of particles engineered to achieve a particular resulting size distribution. But a terminology referring simply to "solar geoengineering by accumulation-mode sulfate aerosols" could lead to some readers inferring too simplified a relationship between the size distribution at particle emission and the evolving size distribution of the resulting dispersed aerosol cloud.

The authors refer to Pierce et al. (2010) and whilst the 2D-AER interactive stratospheric aerosol simulations give a reasonable assessment for the progression of the geoengineered aerosol cloud, the dilution of the initial plume and its subsequent evolution of the size distribution of the dispersed aerosol within the stratospheric dynamics of a higher resolution 3D GCM may well have given differing result.

As I say, I am not at all underplaying the value of these model experiments, which could well help to shed light on some of these issues, but I strongly advise the authors use a different terminology for the mechanism the model experiments are investigating.

Within the article, the authors need to be clearer whether these experiments really are representing a scenario of injected H2SO4 vapour. With the resulting plume rapidly nucleating particles to form a source of new particles that progress to be large enough to scatter incoming solar radiation.

The current model experiments do not really represent that situation, because there would certainly be greater variability in the size distribution in that case of H2SO4 vapour emission. Rather I would argue these experiments mimic a situation where particles are emitted with a controlled size distribution, the particles deliberately engineered to achieve a certain subsequent response within the stratospheric aerosol layer.

My recommendation in this first comment is to change to a more general title something like:

"A co-ordinated intercomparison of interactive stratospheric aerosol model experiments for hypothesised scenarios of solar radiation management by sulfate aerosols"

That's partly because the experiments are not restricted to only assess an emitted source of particles, they also assess the models' response to emitted SO2. In light also of the potentially large variations in particle size distribution that would result, to simply tag the approach as "Accumulation mode particle geoengineering" is not appropriate (in my opinion).

As per the subsequent specific comments, within the article, I can understand there is a benefit to referring to the effect from the strategy (in that it is specifically introducing accumulation mode particles into the models), but still I'm recommending the authors use a different terminology than "AM-H2SO4 geoengineering".

Global aerosol microphysics modellers may tend to use the term "sub-grid scale particle formation" or "primary sulphate emission" for this approach, with the former being much preferred to the latter by experts. And in my comments then I advise to use the term "sub-grid scale particle formation model experiments" or similar as the alternative term.

> *We prefer to retain the term "AM-H2SO4" as it is less wordy than alternatives. However, we add more description of these scenarios to avoid oversimplification and misleading readers.*

2) Page 1 -- Abstract, line 11 -- Suggest to insert "tended to focus on" rather than simply "focussed on", and rather than the somewhat vague term "Analyses", be clear you're referring to interactive stratospheric aerosol model analyses". In fact probably better to use "studies" rather than "analyses".

> *Changed first part of sentence to read "Model studies of stratospheric solar geoengineering have tended to focus on sulfate aerosol enhancement..."*

3) Page 1 -- Abstract, lines 11-12 -- the 2nd half of this 1st sentence then refers to climate models (whereas I think the first half refers to interactive stratospheric aerosol models). I think I understand what the authors mean when they say the climate model experiments "have assumed injection of SO2", but that could confuse some readers, because the majority of climate models do not tend to use their interactive aerosol modules for stratospheric aerosol, and therefore do not tend to represent injection of SO2 at all.

I think what the authors mean is that the model experiments tend to be designed to represent a scenario of imposing radiative effects consistent with best estimates of what could be expected from continued injection of SO2.

I suggest to change "have assumed injection of SO2" to "are based on scenarios aimed to represent the effects from continued SO2 injection". Or similar.

> *Changed this part of sentence to: "and almost all such climate model experiments are base on scenarios which assume injection of SO_2 for this purpose."*

4) Page 1 -- Abstract, line 12 -- As per my general comments above, "may produce overly large aerosols" is obviously unscientific language. Also, that particles grow larger with increased SO2 is a scientific fact, with then use of the word "Yet" not good grammar. It's an important point the authors are making, but this should be stated in an objective way, whereas the precursor word "Yet" suggests the authors consider it somehow unfortunate or undesirable.

Suggest "It is well established (e.g. Pinto et al., 1989) that greater emission of SO2 leads to larger sulphate aerosol particles, with shorter residence time in the stratosphere."

> *Suggested wording adopted and used in the introduction, to avoid references in the abstract.*

5) Page 1 -- Abstract, line 13 -- I think changing "new" to "additional" changes to a more accurate representation, to ensure authors do not mistakenly infer that particles form immediately at accumulation mode sizes (but rather grow from an initially smaller Aitken mode sizes) in this scenario of aircraft injection of SO3 or H2SO4.

This is an example of where I think the simplified term "geoengineering by accumulation mode sulphate" might only increase the probability of an incorrect inference in that respect. I therefore strongly suggest the authors delete "AM-H2SO4", as the acronym similarly will tend to embed an increased likelihood of that over-simplified perception of the progression of the microphysical and dynamical processes involved.

The term "nudged" is also not appropriate in this context, tending to over-simplify the response of the stratospheric aerosol layer.

I'd suggest to re-word to "Some studies have explored whether a stream of very small particles can be generated by injecting H2SO4 vapour rather than SO2, potentially then leading to longer-lived aerosol particles for a given sulphur injection rate."

Introducing a specific delivery mechanism seems un-necessary, and my suggested re-wording then also keeps the point more general than that specific situation of aircraft injection.

> *We have modified the abstract to read: "Injection of SO_3 or H_2SO_4 from an aircraft in stratospheric flight is expected to produce additional accumulation-mode particles (AM-H_2SO_4) after microphysical processing within an expanding plume, and such injection may allow the resulting stratospheric sulfate aerosol layer to more effectively scatter solar radiation."*

6) Page 1 -- Abstract, line 15 -- For the reasons given earlier, please change the terminology "AM-H2SO4 injection" to refer to the specifics of the model experiments rather than an apparently more general "type of geoengineering". As

explained in my comments above, I'm suggesting to use the term "sub-grid scale source of particles" as the descriptor, referring then to the specifics of the model experimetns, with also an acronym then not required in this case.

> *We prefer to retain AM-H2SO4 but have clarified the term and it's implicit sub-grid processing: "Injection of SO_3 or H_2SO_4 from an aircraft in stratospheric flight is expected to produce additional accumulation-mode particles (AM-H_2SO_4) after microphysical processing within an expanding plume, and such injection may allow the resulting stratospheric sulfate aerosol size distribution to more effectively scatter solar radiation. . We report the first multi-model intercomparison to evaluate the effects of such an approach, which we label AM-H_2SO_4 injection based on the size distribution input to global-scale models after implicit sub-grid processing."*

I suggest then to change this sentence to instead say "Whereas GeoMIP has included experiments to intercompare SO2 injection scenarios, the results here are the first multi-model intercomparison of the effects from a sub-grid scale source of sulphate aerosol. Or something like this.

With the subsequent sentence referring to GeoMIP, suggest to reserve the statement of "first" for after the sentence referring to GeoMIP. The scope of that sentence can be made more general by changing "We compare three models" to "A co-ordinated multi-model experiment designed to represent this SO3- or H2SO4-driven geoengineering scenario was carried out with 3 interactive stratospheric aerosol models:". The word "coordinated" can then be deleted later in the sentence.

> *"We report the first multi-model intercomparison to evaluate and compare the effects of such an approach, which we label AM-H_2SO_4 injection based on the size distribution input to global-scale models after implicit sub-grid processing. A co-ordinated multi-model experiment designed to represent this SO_3- or H_2SO_4-driven geoengineering scenario was carried out with 3 interactive stratospheric aerosol-climate models…"*

7) Page 1 -- Abstract, line 24 -- Further to my general comments above, the term "sensitivity to injection pattern" is not an adequate description of the two experiments, the two-site experiment resulting in a midlatitude-focused forcing with little enhancement to the tropical stratospheric reservoir. The word "sensitivity" suggests a slight change whereas these two alternative representations of the geoengineering enhancement are much more substantially different. Better to actually crystallise in the reader's mind what the two alternative scenarios represent -- a mid-latitude-focussed forcing (presumably designed to avoid perturbing climate-sensitive regions in the tropics?) and an evenly distributed injection rate across the tropics and mid-latitudes.

Suggest then to change the sentence beginning "We explore the sensitivity to injection pattern" to "Simulations with two scenarios were designed to compare a two-site injection focused to force only the mid-latitudes, with a more evenly distributed geoengineering forcing, each run with both SO2 and sub-grid particle experiments." or something like this.

The "and find opposite impacts" is explaining the results, and should be explained in a separate sentence, changing "and find opposite" to "We find opposite" or similar.

> *We have revised this part of the abstract to read: "We use two different injection patterns, one designed to force mainly the midlatitudes by injecting at 30° N and 30° S and the other designed to force more uniformly by injecting in a belt along the equator between 30° S and 30° N. By forcing each case with both SO_2 and AM-H_2SO_4, we find opposite impacts on radiative efficacy for the two injection patterns, suggesting that prior model results for concentrated injection of SO_2 may be strongly dependent on model resolution."*

8) Page 2 -- Introduction, line 2 -- insert "long-wave" before "radiative forcing" and change "from the rise in CO2" to "from increased CO2 concentrations". The reason here is to keep in the reader's mind that emissions are not necessarily the same as concentrations.

> *Suggested language adopted.*

9) Page 2 -- Introduction, line 3 -- "Despite the complexity" -- it's not corect to say "Despite" here and I'd argue it's more "because of the complexity" that these models are needing to be used to try to predict how the overall system responds given the complex interactions and feedbacks.

Suggest to change "Despite the" to "In light of the".

> *Suggested language adopted.*

10) Page 2 -- Introduction, line 4 -- "solar radiation management (SRM) is being studied". It's not really the solar radiation management itself that is being studied -- it's the effects from hypothesized solar radiation management (whether that be the responses of the stratospheric aerosol and ozone layers or the surface response to climate and the hydrological cycle).

Suggest to insert "the effects from hypothesized" after "carried risks".

> *Suggested language adopted.*

11) Page 2 -- line 5 -- since the word "climate" is used later in the sentence (and with the change above also earlier in the sentence) change "climate models" to "earth system models". And add citations to 2 or 3 of the key papers here.

> *Suggested language adopted. Full sentence now reads:* "In light of the complexity of the climate system and the inherent risks of climate manipulation, the effects of hypothesized solar radiation modification (SRM) are being studied with earth system models to examine the potential benefits and possible adverse effects (e.g. Aquila et al., 2014; Richter et al., 2017; Tilmes e al., 2017) while simultaneously improving our knowledge of climate interactions and feedback processes."

12) Page 2 -- line 11 -- change "the climate response to stratospheric aerosol injection" to "the climate response to a geoengineering-enhanced stratospheric aerosol layer" or similar.

It's the eventual enhancement to the stratospheric aerosol layer that causes the forcing, not the injection. With a residence time of months to years, there is quite some difference between the nature of any injection and the resulting forcing that the climate then responds to.

Suggested language adopted.

13) Page 2 -- line 13 -- change "alter the climate" to "cool the surface climate and warm the stratosphere"

Suggested language adopted.

14) Page 2 -- line 19 -- change "Studies of SSG" to "Studies of SRM" -- and change other instances of "SSG" within the paper instead to "SRM"

Suggestion adopted.

15) Page 2 -- line 20 -- change "unfavorable size distributions" to "shorter residence time in the stratosphere (larger particles)".

Changed "unfavorable size distributions" to "larger particles (less efficient shortwave scattering) and shortened aerosol residence time"

"16) Page 2 -- line 25 -- As I explained in my general comments, "These limitations might be addressed.." is not scientific language, and should be focused on which of the 5 limitations stated, emitted "various solid particles" the suggested solid particles is intended to address.

Changed this to "Limitations (3) and (4) might be addressed through use of various solid aerosol particles for SRM"

17) Page 2 -- line 25 -- The phrase "altering the size distribution" does not adequately communicate the complexity of the microphysical and dynamical processes that combine to effect the stratospheric aerosol layer's adjustment to a geoengineering source of aerosol particles. Whilst I understand that a strategy can be designed for engineered particles to aim to achieve a given desired size within the subsequent months to years of their circulation within the stratosphere, it is over-simplifying this to refer to "altering the size distribution". It is of course certainly possible to alter the size distribution of the emitted particles, but any control within the response of the stratospheric aerosol layer in the subsequent months is too uncertain to be referred to simply as "altering the size distribution".

Please change "alternatively some of the limits may be addressed by altering the size distribution" to "with engineering strategies potentially able to achieve a more prolonged aerosol particle residence time in the stratosphere". Or if the authors mean the radiative efficacy, please phrase this more explicitly to be clear of the size effect intended.

> *Changed to "alternatively limitation (1) may be addressed with geoengineering strategies designed to achieve a sulfate aerosol layer with a size distribution that optimizes shortwave scattering"*

18) Page 2 -- line 28 -- The sentence beginning "Efficacy is decreased" needs to be re-written for at least two reasons. Firstly, this is the first time the word "Efficacy" has been introduced, and it's not clear where this is scattering efficiency or efficacy in terms of residence time. The remainder of the sentence suggests it is mainly the latter -- so rather than "Efficacy is decreased...", suggest instead to say "Aerosol particle residence time in the stratosphere reduces...".

> *Suggestion adopted.*

19) Page 3 -- line 15 -- "Our aerosol size distribution" is unscientific language. Please change to "The constant size distribution used by the models in the co-ordinated experiment..."

> *Suggestion adopted.*

References

Dyer, A. J. and Hicks, B. B. "Global spread of volcanic dust from the Bali eruption of 1963" Q. J. Roy. Meteorol. Soc., vol. 94, pp. 545-554, 1968.

Dyer, A. J. "The effects of volcanic eruptions on global turbidity, and an attempt to detect long-term trends due to man", Q. J. Roy. Meteorol. Soc., vol. 100, pp. 563-571, 1974.

Grant, W. B., Browell, E. V., Long, C. S., Stowe, L. L., Grainger, R. G. and Lambert, A.: "Use of volcanic aerosols to study the tropical stratospheric reservoir" J. Geophys. Res., vol. 101, no. D2, pp. 3973-3988, 1996.

Pinto, J. P., Turco, R. P. and Toon, O. B.: "Self-limiting physical and chemical effects in volcanic eruption clouds" J. Geophys. Res., vol. 94, no. D8, pp. 11,165-11,174, 1989.

> *Three of these four references have been included in the revised manuscript*.

Response to Reviewer 3

We thank the reviewer for careful reading of our manuscript and constructive comments. Our responses to the reviewer's general and specific comments are given below in italics following each comment. Considering the issues raised by the reviewer has allowed us to improve our manuscript by clarifying these issues in the text. We are grateful for the reviewer's time and thoughtfulness.

- This study investigates the implications of using SO_3 or H_2SO_4 instead of SO_2 in deliberate emissions in the stratosphere in order to modify Earth's climate. Using SO_3 or H_2SO_4 would produce smaller particles (accumulation mode – AM-H_2SO_4) which are more radiative effective than those formed from emissions of SO_2. The effects of geoengineering with AM-H_2SO_4 is investigated using three global climate models. The effects on the stratospheric size distribution, aerosol load, temperature, water vapour and ozone as well as the radiative effects are investigated. All models show that there is increased radiative efficiency using AM-H_2SO_4 but there are large intermodel differces.

  The study is well performed and many different aspects of using AM-H_2SO_4 instead of SO_2 is investigated. This type of investigation using three models in one study has not been performed before. The three models used in the study have different strength and weaknesses in their representation of the stratosphere which gives relevant information of the uncertainties in the modelling geoengineering in the stratosphere with AM-H_2SO_4 and SO_2. The paper is well written in general and has a clear structure. The paper is well within the scope of ACP and I recommend publication after the following comments has been addressed.

  General comments:

  It would be interesting to include a short discussion on the feasibility of using SO_3 or H_2SO_4 instead of SO_2 and whether one of the options is more technically challenging than the other one.

  We have added a sentence regarding the technical and engineering challenge of using SO3 or H2SO4 and included two references, Smith et al., 2018 and Janssens et al., 2020.

  Specific comments:

  Page 6, line 27: Why were the emissions released at different heights in the different models?

  This was a function of the model's vertical grid resolution and necessary conversion from altitude to pressure level.

  Page 6, line 30: I miss an explanation or motivation of the choice of the different injections and injections points. What was the scientific motive for choosing those emissions and emissions points? Which scientific questions could be answered with these?

  We added more explanation of the injection patterns in the scenario descriptions in Section 2: "The regional injections are designed to utilize the Brewer-Dobson circulation to distribute emissions globally and maximize their residence time, as has been observed for volcanic aerosol clouds (Dyer, 1968; Grant et al., 1996). The 2point injections occur outside the tropical stratospheric reservoir (Grant et al., 1996; Tilmes er al., 2017) and are meant to concentrate geoengineering impacts at higher latitudes and to explore microphysical differences when injections are more concentrated spatially."

Page 12, line 26: “main particle size distribution from an R_g”. What is the main size distribution R_g? R_g was defined as the mode radii value, but the main size distribution cannot have one mode radii value.

Corrected to be the accumulation mode.

Page 24, line 11-16. There is quite a lot of discussion here that has not been included previously in the manuscript. The section head should perhaps be changed from “summary and conclusion” to “summary and discussion.”

Adopting this suggestion.

Technical corrections:

Page 9, line 7: It is a bit vauge to start the sentence with ”This figure” no figure has been mentioned for several sentences.

Replaced “This figure” with “Figure 2”.

Page 10*, line 10-14. This sentence is very long. Please divide it.*

Done.

Page 13, line 7: This sentence is awkward, please revise.

Revised to read: “The size distributions respond differently to 2point rather than regional injections depending on whether SO_2 gas or AM-H_2SO_4 particulate is injected. These results suggest the way aerosol microphysics drives differences between AM-H_2SO_4 and SO_2 injection scenarios (see Table 3).”

Figure 11: The legend in this figure uses SO_2 and H_2SO_4 to denote the simulations rather than AM-H_2SO_4as in the rest of the manuscript. Please revise for consistency.

Corrected.