

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 distributions 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 surrounding 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 alternative 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:

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 based on scenarios which assume injection of SO₂ 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 SO₃ or H₂SO₄ from an aircraft in stratospheric flight is expected to produce additional accumulation-mode particles (AM-H₂SO₄) 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 experiments, 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 SO₃ or H₂SO₄ from an aircraft in stratospheric flight is expected to produce additional accumulation-mode particles (AM-H₂SO₄) 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-H₂SO₄ 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-H₂SO₄ 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 SO₃- or H₂SO₄-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 SO₂ and AM-H₂SO₄, we find opposite impacts on radiative efficacy for the two injection patterns, suggesting that prior model results for concentrated injection of SO₂ 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 correct 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 et 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 coordinated experiment..."

Suggestion adopted.

References

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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.

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