Report #2 by Reviewer #2

We thank the review for another careful reading of our manuscript. His/her comments have helped us clarify several points and improve the manuscript. In two cases the reviewer misinterpreted our meaning and therefore we have clarified the text and stated our meaning more explicitly. In other cases we have adopted the reviewer's advise and appreciate the rewording suggestions. Our specific replies to reviewer comments 1 through 6 are listed below in italics and green text.

Submitted on 29 Dec 2021
Anonymous Referee #2

Anonymous during peer-review: **Yes** No

Anonymous in acknowledgements of published article: **Yes** No

Recommendation to the editor

1) Scientific significance Does the manuscript represent a substantial contribution to scientific progress within the scope of this journal (substantial new concepts, ideas, methods, or data)?	Outstanding **Excellent** Good Fair Low
2) Scientific quality Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way (consideration of related work, including appropriate references)?	Outstanding **Excellent** Good Fair Low
3) Presentation quality Are the scientific results and conclusions presented in a clear, concise, and well structured way (number and quality of figures/tables, appropriate use of English language)?	Outstanding Excellent Good **Fair** Low

For final publication, the manuscript should be

accepted as is

accepted subject to **technical corrections**

accepted subject to minor revisions

reconsidered after **major revisions**

rejected

Were a revised manuscript to be sent for another round of reviews:

I would be willing to review the revised manuscript.

I would not be willing to review the revised manuscript.

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

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

I was one of the 3 reviewers who reviewed the submitted version of this manuscript in August 2021, the analysis presenting 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: one emitting only at 30N and 30S, the other as a constant source between 30S and 30N.

As I identified in my original review, the intercomparison (across 3 different models), 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.

My original review explained the analysis would be publishable in a revised form, my review found the aim and design of the model experiments to be poorly described, with the Introduction and interpretation needing to include some discussion also of the tropical stratospheric reservoir, in relation to expected differences between the two scenarios.
I also noted several places where the manuscript had unscientific language or vague statement, and a few places where the wording was poor, or over-simplifying the changes to the stratospheric aerosol layer that would occur in this hypothesised large-scale injection of precursor gas or idealised particle for solar radiation management.

I made a list of 20 specific revisions that were needed, before the manuscript could potentially then proceed to publication, with also a request that the authors change the title, and querying the terminology "Solar Geoengineering by Accumulation-Mode Sulfate Aerosols", in being inconsistent with what the authors state that particular model experiment is representing.

With some of the comments relatively fundamental to the narrative of the manuscript, I found that major revisions were needed, but with the Figures much of the results section is in good shape, these were relatively minor revisions.

The authors have replied positively to each of the specific comments I made, with the instances of imprecise wording in the manuscript now remedied, with also most of the instances where particle size changes were over-simplified now also much improved.

One of my main comments was to advise the authors to change the term "geoengineering by accumulation-mode sulfate aerosols" to simply "geoengineering by sulfate aerosols.
This suggestion was on the basis that that terminology also somehow suggests a relative ease of achieving that particular desired size, or that the particles will remain at a particular "target size" in their months to years lifetime in the stratosphere.

As I explained in my review, the vertical and meridional variations in stratospheric aerosol particle size seen in the years after the Pinatubo eruption, and other major eruptions, and from my own experience from analysing a range of major eruption scenario experiments within interactive stratospheric aerosol models suggest the range of variation in particle size would likely be broader than many readers would infer from that "geoengineering by accumulation-mode sulfate aerosols" terminology. Somehow that terminology suggests, to me

at least, is that it is a relatively simple issue to achieve the required particle size.

In their reply to this comment, the authors explain that their preference to retain that same terminology -- and of course this is their manuscript -- and given the improvements in the wording the authors have made, despite my continued opposition to that term (it still communicated an over-simplified situation), it is OK for that terminology to remain in place within this particular article.

Although the manuscript is much improved, there are still some places where the wording requires minor changes, to then be fine to proceed to publication. These remaining few minor revisions are listed below, with 2 of the 6 required changes being more substantial than the others, and requiring some explanation here.

Both of these two remaining "substantive points" relate to the way the text in the revised manuscript refers to the specific type/class of models whose results are analysed in this multi-model analysis.

The first of the 2 is to ask the authors to change the name they have used for the type/class of models used in the analysis, specifically within the new title the authors have added in the revised manuscript, where the models are referred to as "aerosol-climate models".

Whilst I accept that the models can generally be referred to within that broad class of models that include the radiative effects of aerosol in their predictions of the earth's climate, the functionality being applied for this analysis, requires a particular capability for "interactive stratospheric aerosol". As the authors will be aware, there is currently ongoing a model intercomparison project/iniative "ISA-MIP" (Timmreck et al., 2018) which has designed model experiments to specifically inter-compare these interactive stratospheric aerosol models, including more background (volcanically quiescent) conditions, and experiments for major eruptions, and to hindcast predictions through the series of more moderate stratospheric-injecting eruptions that have occurred so far in the 21st century.

For that MIP, the terminology the community agreed for these models was "interactive stratospheric aerosol models" (the ISA within ISA-MIP) and whereas these models are certainly aerosol-climate models, the interactive stratospheric aerosol capability is key here in terms of being able to predict the onward variations in particle size, and associated residence time, from the initial "emission size distribution" the particles initially as they mixed into the ambient air around the aircraft.

And that 1st of the 6 changes is then requesting to change the title from "An aerosol-climate model intercomparison" instead to "An interactive stratospheric aerosol model intercomparison".

We have modified the title as suggested, using the terminology adopted for ISA-MIP.

It is notable to me that each of the 3 interactive stratospheric aerosol models compared here also having the particular sophistication to represent microphysical processes within their interactive predictions of the stratopheric aerosol layer's variations. And then I am advising here to also add the word "microphysics" before "intercomparison" -- this then being an "interactive stratospheric aerosol microphysics intercomparison. I leave it up to the authors however to choose either of those -- each an improvement on the much less specific "aerosol-climate model" terminology, which is too broad to adequately communicate the particular type of experiments the manuscript analyses results from.

The other of the 2 remaining substantive change also relates to the type of model, where I think the authors current description of the "modus operandi" of the models needs changing.

Specifically the wording:

"As input, they would take the particle size distribution from aircraft plume model studies but could represent any hypothetical input of particles".

This reference to an "input size distribution" under-plays the value of the interactive stratospheric aeorsol models, and could lead some readers to mis-understand the aerosol-climate models to simply be enacting a "prescribed but globally varying" size distribution from some other experiments with an interactive stratospheric aerosol model. This is not the case, with the interactive models doing much more than simply representing a particular "input size distribution".

Each of the 3 interactive stratospheric aerosol microphysics models used in the study predict how the initially localised plume of geoengineering aerosol would progress to the "response" of a global enhancement to the stratospheric aerosol layer, and how that would evolve in the months of years of the continued injection/emission.

The global spatial variation of the particle size, and its temporal variability across different seasons and years, with the internal variations in the stratospheric circulation and its dynamical states that occur through the simulations. The relationship between a particular engineered particle size at emission, and the eventual variation in size one would see at a global scale (across the tropics, midlatitudes and high latitudes of each hemisphere) is far from certain. In addition to the transport variations, the coagulation of the particles, and the subsequent removal and vertical distribution from gravitational settling, will likely cause substantial variations in the progressions in particle sizes that would occur as the continuing (or intermittent) plume(s) are dispersed globally over months and years, would introduce subsequent variations that differ from the initial "emission size distribution" introduced from the emission location (e.g. aircraft or tethered pipe).

This issue communicates a bit more about the basis of my objection to my the "geoengineering by accumulation-mode sulphate" terminology, in some readers inferring an over-simplified situation in how the dispersed particles would "end up" within the "enhanced state" of a geoengineered stratospheric aerosol layer.

Whilst I am content to concede that to the authors choice of terminology, when referring to the models being used in the analysis, the text needs to better communicate what the interactive stratospheric aerosol models actually represent. In particular the reference to an "input size distribution", seems to be an over-simplfication.

We realize the confusion between "input size" and global aerosol distribution. Therefore we have changed the wording to refer to a "mass flux of particles" or "source of particles at a specified rate". This should clarify the difference between the input particle sizes and the resulting global aerosol distribution. On page 4, lines 3-4 now read: "As input, the global models would take a mass flux of particles with the size distributions generated by aircraft plume model studies or any hypothetical source of particles at a specified rate."

The other 4 changes are more minor, and self-explanatory, and I then list below the 6 minor revisions which I'm advising are required before the manuscript can then proceed to publication in ACP.

Remaining minor revisions
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1) Manuscript title: Page 1, line 1 -- please change "An aerosol-climate model intercomparison" to either "An interactive stratospheric aerosol model intercomparison" or preferably (from my perspective)
"An interactive stratospheric aerosol microphysics model intercomparison". I note that the authors refer to "interactive stratospheric aerosol-climate models" (e.g. page 1, lines 20-21), and a potential variant of that 2nd suggested alternative could be to change the words "aerosol microphysics" for "aerosol-climate".

We have adopted "An interactive stratospheric aerosol model intercomparison of solar geoengineering by stratospheric injection of SO_2 or accumulation-mode sulfuric acid aerosols" as the title. In lines 20-21 of the abstract we replace" aerosol-climate models" with "aerosol microphysics models".

2) Abstract: Page 1, lines 12-14 -- this 1st line of the Abstract still seems poorly worded to me. I don't understand what the authors are trying to communicate here. The wording currently states:

"Studies of stratospheric solar geoengineering have tended to focus on sulfuric acid aerosols, and almost all such climate model experiments assume that SO2 is injected to increase the sulfuric acid aerosol burden of the stratosphere."

I think this sentence should be replaced with a more meaningful comment on previous work, in relation to the difference between imposing a particular particle size, and simulating the size distribution interactively. This paper is the 1st ever inter-comparison of interactive stratospheric aerosol simulations of the geoengineering-enhanced stratospheric aerosol layer, and it is this functionality that I'd suggest this functionality the first sentence of the Abstract focuses on communicating.

A specific suggestion would be to replace that sentence with:

"Previous model comparisons of stratospheric solar geoengineering have mostly tended to focus on climate model experiments assuming a particular prescribed particle size for geoengineered sulfate aerosol particles."

The 1st part of the 2nd sentence of the Abstract should also be improved -- as it is similarly poorly worded at present -- my specific recommendation is to re-word that 1st part to instead begin "By contrast,
a key finding from interactive modeling studies is that the radiative forcing of a geoengineered stratospheric aerosol layer would increase sub-linearly....."

The review misunderstands the meaning and intent of the first sentence of the abstract. We state that most studies of stratospheric solar geoengineering have focused on sulfuric acid aerosols (as opposed to solid particle injections) and that most of these studies (other than those that simply modify the solar constant or impose a stratospheric size distribution) assume injection of SO_2. The wording was not clear. We have made the wording in the first 3 sentences of the abstract much more precise:

"Studies of stratospheric solar geoengineering have tended to focus on modification of the sulfuric acid aerosol layer, and almost all climate model experiments that mechanistically increase the sulfuric acid aerosol burden assume injection of SO_2. A key finding from these model studies is that the radiative forcing would increase sub-linearly with increasing SO_2 injection because most of the added sulfur increases the mass of existing particles, resulting in shorter aerosol residence times and aerosols that are above the optimal size for scattering. Injection of SO_3 or H_2SO_4 from an aircraft in stratospheric flight is expected to produce particles predominantly in the accumulation-mode size range following microphysical processing within an expanding plume, and such injection may result in a smaller average stratospheric particle size, allowing a given injection of sulfur to produce more radiative forcing.."

3) Abstract: Page 1, lines 28-30 -- The "We use" wording here is colloqial and needs to be changed, with also the "injection patterns" and "belt" somehow (to me at least) not sufficiently communicating the scientific issue being explored. The experiment that injects at 30N and 30S is presumably a specific "deployment scenario", aimed to force only the mid-latitudes, whereas the constant emissions from 30S-30N
is a more theoretical scenario, perhaps idealsed to achieving the longest residence time for the emitted particles. Suggest to re-word:

"We use two different injection patterns"

instead to

"The models carried out two different "geoengineering-enhancement scenarios" or similar more scientific terminology than "patterns".

I think there is an error here also where you state "injecting in a belt along the equator between 30S and 30N" --- perhaps it's simply a case of deleting "along the equator", but I'd recommend also changing "in a belt" to "uniformly" or "with a uniform emission rate" or similar.

The wording here did need improvement. We replace "injection pattern" with "geographical distributions of injection mass", and add "idealized" in front of "geographical distribution". We also change "belt" to "region". And further clarify the 30S-30N injection to be "uniformly in the regiom between 30S and 30N" and motivated to "maximize aerosol residence time". The new working is:

"The model studies were carried out with two different idealized geographical distributions of injection mass representing deployment scenarios with different objectives, one designed to force mainly the midlatitudes by injecting into two grid points at 30° N and 30° S and the other designed to maximize aerosol residence time by injecting uniformly in the region between 30° S and 30° N."

4) Abstract: Page 1, lines 30-32 -- This last sentence of the Abstract also needs to be improved, as it's not clear what is meant by "opposite impacts" -- and the term "radiative efficacy" seems somehow ill-defined (or not yet introduced). Perhaps a simple re-wording of "opposite impacts on radiative efficacy" to "strongly differing radiative forcing efficacy" -- or just "strongly differing radiative forcing".

We agree that this sentence was not clear and that radiative efficacy is not yet defined in this context. Therefore we change this sentence to make our point about concentrated (2point) vs dispersed (regional) injection with reference to size distributions instead: "Analysis of aerosol size distributions in the perturbed stratosphere of the models shows that particle sizes evolve differently in response to concentrated versus dispersed injections depending on the form of the injected sulfur (SO_2 gas or AM-H_2SO_4 particulate) and suggests that prior model results for concentrated injection of SO_2 may be strongly dependent on model resolution" This concept is explained in Table 3 and we feel it should be highlighted in the abstract.

5) Introduction: Page 3, lines 27-30 -- The current wording "Detailed modelling of this complex process for a full range of stratospheric physical, chemical and microphysical conditions awaits further studies" should be improved to better communicate the scientific issue here (rather than the technical aspects of the processes involved). I mean the question of how the particle size progresses, over a timescale of months to years, from the initial size at the plume-scale, to the global-scale variations in particle size in the "dispersed state" of the geoengineering-enhanced stratospheric aerosol layer.

Suggest instead "A priority for future modeling studies could potentially be to establish how the initial "engineered particle size" at the plume-scale progresses to the global-scale variations in particle size in the "dispersed state" of the geoengineering-enhanced stratospheric aerosol layer."

This sentence refers to plume evolution, not large-scale evolution. We have revised it to now read: "Detailed modelling of potential plume-scale evolution under a full range of stratospheric physical, chemical, and microphysical conditions awaits further studies."

The follow-on sentence should also be re-worded. The current text refers to "the GeoMIP models" but, further to my general comments above, I think the authors mean the interactive stratospheric aerosol
models -- i.e. "the ISA-MIP models". Also change "can be used to simulate" to "have the functionality to
explore how the stratospheric aerosol layer responds with the global dispersion of the geoengineering particles." Or something like this.

We have added the reference to Timmreck et al., 2018 and modify this sentence as suggested: "For the temporal and spatial scale beyond plume models, global GCMs such as those participating in the Interactive Stratospheric Aerosol Model Intercomparison Project (ISA-MIP, Timmreck et al., 2018) have the functionality to explore how the stratospheric aerosol layer responds with global dispersion of the geoengineering injections." In the next paragraph, after naming the three models participating in this study, we add: " These three models are participants in both the GeoMIP and ISA-MIP model intercomparisons."

6) Introduction: Page 3, lines 30-33 -- The 1st of the 2 setnences here refers to ozone, temperature and circulation -- but the impacts on ozone depends strongly on the stratospheric chemistry scheme, which is outside the scope of this article. I suggest to narrow the scope of this sentence to instead to focus on the aerosol changes. A specific suggestion is to change:

"These GCMs can effectively simulate changes in global aerosol
burden, radiative forcing, ozone, and stratospheric temperature and circulation

instead to

"Those ISA-MIP models with microphysical aerosol schemes can also address the key issue of how the particle size distribution progressees, this being a key determinant of subsequent global aerosol burden and radiative forcing."

As in my general/overarching comments above, the 2nd sentence here also needs changing, with this issue of an "input size distribution" needing to be better explained.
The wording says:

"As input, they would take the particle size distribution from aircraft plume model studies
but could represent any hypothetical input of particles".

I think this reference to an "input size distribution" could be mis-interpreted by some readers unfamiliar with the types of model involved. The value of the interactive stratospheric aerosol models is more than simply that they can represent an input size from a plume-scale model. It's this issue of how the emitted particles subsequently transform in a "globally dispersed state", as they become part of a geoengineering-enhanced stratospheric aerosol layer.

We adopt much of the suggested rewording in this paragraph. "Those models with microphysical aerosol schemes can also address the key issue of how the particle size distribution evolves, this being a key determinant of subsequent global aerosol burden and radiative forcing. As input, the global models would take a mass flux of particles with the size distributions generated by aircraft plume model studies, or any hypothetical source of particles at a specified rate. The input size distribution is simplified here by using a lognormal distribution with a constant mode radius

and mode width for all injection grid points and times."

By clarifying that the geoengineering particle injections involve both a size distribution and a mass flux rate, we hope to avoid misinterpretation that a size distribution is imposed on the global stratosphere.

With the re-wordings in points 1 to 5, it may be that this is then sufficiently explained, with then this final sentence explaining of the potential for future work to involve a combination of plume-scale models and the global interactive stratospheric aerosol GCMs.

A specific suggestion for re-wording could be:

"There is the potential for future interactive stratospheric aerosol model experiemnts to link directly with plume-scale model experiments, and seek to realistically represent potential alternative deployment scenarios".

We have added this statement to the last paragraph of the summary and discussion section which mentioned plume-in-grid and adaptive mesh as methods of combining plume and global scale modeling.
"Improved understanding of the effectiveness of stratospheric sulfur injection and the role of plume-scale formation of accumulation mode particles may require use of modelling methods such as plume-in-grid or adaptive mesh to better capture the multi-scale problem from injection plume to the global circulation. Such methods may allow future interactive stratospheric aerosol model experiments to link directly with plume-scale model experiments, and seek to realistically represent potential alternative deployment scenarios."

References

Timmreck, C., Mann, G.W., Aquila, V., Hommel. R., Lee, L. A., Schmidt, A. et al. (2018):
"The Interactive Stratospheric Aerosol Model Intercomparison
Project (ISA-MIP): motivation and experimental design"
Geosci. Model Dev., 11, 2581–2608.
https://doi.org/10.5194/gmd-11-2581-2018

Reference added.