Response to reviews of:

Solar geoengineering using solid aerosol in the stratosphere
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ACPD-15-11799

Response to Anonymous Referee #1

Received and published: 12 May 2015

> First we would like to thank the reviewer for a careful reading of our manuscript and many constructive comments that have improved the quality and clarity of the paper. We have added a table to list the model experiments described and an appendix to detail the coagulation and condensation schemes. We added a paragraph on the limitations and inherent size broadening of a discrete sectional bin scheme. We have included a more thorough description of the AER 2-D model's dynamical fields and PSC parameterization and discuss some model comparisons with observations for both aerosols and ozone. We adjusted our RF values in Figures 7 and 13 due to a correction in the albedo adjustment. We added a comparison of ozone change from sulfate geoengineering to that from alumina and diamond geoengineering in Figure 12a. We also replace the unit megatons with terragrams throughout the manuscript. We have added seventeen new references and increased font size and readability of the figures. J. A. Dykema has been added as a co-author (previously recognized in a footnote and acknowledgement) due to his scientific contributions in addressing reviewer questions regarding heating rates and radiative forcing.
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> Our answers to the reviewer's questions and comments are detailed below, with the reviewer's comments in black and our responses indented and in green.

First I should say I have not evaluated the ozone depletion part of the paper as this falls out of my expertise, and have focused instead on the physical part of the study. I confess I am not very knowledgeable in fractal aerosols and have not checked the details of the authors' treatment in this study.

Independently of what one may think of the idea of injecting alumina particles in the stratosphere, I think this is, scientifically speaking, a good study. The limitations of the model used (simplified injection mechanism and lack of a plume model, 2D modelling, geometry of aggregates, ozone chemistry and missing feedbacks on the stratospheric circulation and ozone distribution) are highlighted and well discussed in Section 4. The conclusions are clearly explained and the results make physically sense.

I have a few major comments:

The number of monomers in a fractal particle is always a power of 2 as a consequence of the sectional representation that doubles the number of monomers in successive bins. Yet fractal particles of different sizes can coagulate and produce particles with any number of monomers. How is this treated in the model? How much of an assumption is this? Actually fairly little is said on the coagulation scheme for coated and uncoated particles. As this is a new development, it would be useful to describe it in an Appendix. Likewise a better description of the other aspects of the scheme like condensation of sulphuric acid on the particles is needed.

> We agree with the reviewer that a more comprehensive description of this aspect of the model was need. We have added Appendix A to explain the coagulation and condensation schemes. We also added a paragraph in section 4.1 to discuss limitations due to aerosol size binning.

It seems that all "radiative forcing" estimates are for SW effects only (at least this is what I understand from page 11816, lines 10-12). If this is the case, this should be made explicit and justified. This said neglecting the LW (positive) radiative forcing is hardly justifiable given that it can vary significantly between the different particle types (as the authors explain themselves).

> Figures 6 and 7 present calculations accounting for only shortwave effects on radiative forcing. We have made this explicit in the text and figure captions. For purposes of geoengineering, we believe shortwave radiative forcing is the most appropriate metric for comparing different geoengineering methods, especially for a model without interactive dynamics. Changes in stratospheric temperature in response to changes in radiative heating rates may lead to more substantial adjustments in longwave radiative forcing relative to shortwave radiative forcing. Thus the instantaneous longwave radiative forcing is a more ambiguous indicator of changes in surface temperature, which geoengineering attempts to offset. This problem is complex and will be dealt with in a future paper by John Dykema of our group. However, we have made a preliminary calculation of the LW vs SW RF and find that the LW is about 10% of the SW for alumina and is negligible for diamond. We have added the following to our discussion of radiative forcing: "We choose to present only shortwave radiative forcing as more relevant to geoengineering intended to offset surface warming after atmospheric adjustments. However, the longwave radiative forcing is only about 10% of the shortwave RF for alumina, though of opposite sign, and is negligible for diamond."

The paragraphs on heating rates are also unclear and possibly incorrect. Heating is caused by both absorption of SW and LW radiation. It seems odd that the authors only consider the latter (at least this is the impression they give). Also the LW heating rates can be positive (heating) or negative (cooling) depending on the aerosol and temperature vertical profiles whereas the authors seem to associate the interactions of aerosols with LW radiation to a systematic heating of the stratosphere. Please clarify.

> Our focus in this paper is on the relative differences between heating rates in the tropical lower stratosphere, where other authors (see Heckendorn et al., 2009) have highlighted the possibility of aerosol heating from geoengineering increasing the stratospheric water vapor concentration. While cooling may result from increasing aerosol concentrations at high latitudes

> (see Ferraro et al., 2011), heating is the result in the tropical lower stratosphere for the aerosols considered here. We have narrowed our discussion in the paper to apply only to the tropical lower stratosphere, and now consider both LW and SW radiation. And we provide the fractional contribution of SW heating to the total heating: "Shortwave heating from alumina is about 15% of the total heating, and from sulfate about 20%. The total heating rate from diamond is almost entirely due to shortwave effects, but is still much less than that for alumina with the same top-of-atmosphere shortwave radiative forcing."

There are a number of notations and units that need to be clarified (as discussed below).

Specific comments:

Page 11800, line 12: sentence is a little unclear (maybe "yet" should read "although") Page 11801, line 5: The study of Ferraro et al (GRL, 2011) could be cited here.

> done

Page 11802, line 7: and also "cirrus formation" if the dynamical effects propagate in the upper troposphere as some models suggest.

> Cirrus effects are too uncertain (both in sign and in magnitude) for specific mention in our introduction, though we did add the italicized words "these risks include, *but are not limited to*," in that paragraph. Potential cirrus effects are dealt with in section 4.1 under "missing feedbacks".

Page 11802, line 11: a citation to Mercado et al (Nature, 2009) or an earlier paper would seem more appropriate here.

> Mercado reference added.

Page 11806, line 3: I am sure the explanation is somewhere in the cited literature, but could you explain why mass is proportional to R^{D_f}. A diagram might help to understand.

> The relationship between R_g and N_i is found to be a statistical

scaling law that holds true over a wide range of R_0 and N_i values, with D_f remaining constant for a given material. This relationship is probably related to the coagulation process. Since N_i is proportional to R_g^{Df}, and total agglomerate mass is just the mass of a monomer times N_i, agglomerate mass is proportional to R_g^{Df}. We have modified text as follows:

> “Larger particles produced by coagulation assume fractal structures that obey a statistical scaling law where the fractal dimension D_f determines how the size of an aggregate of particles is related to the number of primary particles. … Thus particle mass is proportional to R_g^{Df}. The fractal dimension D_f for a given material has been found to be invariant for a wide range of R_0 and N_i values.”

Page 11807, line 9: what is R? it has not be defined previously. Or do you mean R_g?

Changed this R to R_p as it referred to spherical sulfate particles.

Page 11807, line 10: is N the same as N_i defined previously?

Changed N to N_i.

Page 11807, line 11: are you talking about area or surface area projection (as on line 3) here? What is area relevant here?

Surface area projection. Text clarified.

Page 11807: I do not pretend I understand the details of fractal aerosols very well, so it would be useful if the authors point to limitations in their model.

Section 4.1 “Geometry of aggregates, effects of size binning” covers these limitations. We added a paragraph about the effects of size binning.

Page 11810, lines 20-30: does this depend on N_i?

Yes, sedimentation rates depend on N_i. That should be clear from Figure 1 and from the equations for sedimentation rate given in Section 2. We have made no change in text.

Page 11813: I am not sure what the authors mean when they say “fractals never contain more than X monomers’‘. Surely there must be but in (very) low concentrations?

> Modified this discussion to refer to “significant concentrations”.

Page 11816, line 5: the word “significant” is used in a very subjective way here. What is a significant or insignificant amount of diffuse radiation for terrestrial ecosystems?

> We have rewritten this sentence as: “Thus in geoengineering applications, alumina and diamond would scatter radiation back and produce substantially smaller increases in diffuse radiation at the surface than would sulfate particles producing the same change in RF.”

Page 11816, line 19: scattering becomes negligible per unit mass particle, but not per unit particle. Eventually it depends how much of the mass is in this range of monomers.

> We changed the text to read “An aggregate of 16 alumina monomers has negligible scattering per unit mass.”

Page 11816, lines 27-29: you should say this earlier.

> Moved this sentence into the previous paragraph.

Page 11818, lines 8-9: note that IR radiative effects result in both heating / cooling depending on the altitude and aerosol vertical profile considered.

> Our focus in this paper is on the relative differences between heating rates in the tropical lower stratosphere, where other authors (see Heckendorn et al., 2009) have highlighted the possibility of aerosol heating from geoengineering increasing the stratospheric water vapor concentration. Present-day trace gas profiles for the tropics based on reanalysis data are used for our heating rate estimations, along with a uniform aerosol layer between 18 and 23 km in the tropics. While cooling may result from increasing aerosol concentrations at high latitudes (see

Ferraro et al., 2011), heating is the result in the tropical lower stratosphere for the aerosols considered here. We have narrowed our discussion in the paper to apply only to the tropical lower stratosphere. We replace "longwave, or infrared (IR), heating in the stratosphere" with "aerosol heating of the tropical lower stratosphere", as we now include both longwave and shortwave heating in our estimates. We add a reference here to Heckendorn et al. (2009) who determined that the heating of the lower stratosphere was primarily caused by IR effects for sulfate. And we later quantify the contributions of LW and SW heating to our totals.

"Aerosol heating of the tropical lower stratosphere is another potential risk of geoengineering. Heckendorn et al. (2009) investigated this effect and the resulting increase in stratospheric water vapor, primarily caused by longwave heating, for sulfate aerosol. … Shortwave heating from alumina is about 15% of the total heating, and from sulfate about 20%. The total heating rate from diamond is almost entirely due to shortwave effects, but is still much less than that for alumina with the same top-of-atmosphere shortwave radiative forcing."

Pages 11816, 11817 and 11818, line 24: are these Wm^{-2} of net (SW+LW) forcing or SW forcing only?

Shortwave only. We have clarified this in the text.

Page 11840: change "mixing ratio" to "mass mixing ratio" for clarity on panels a) and b). A mixing ratio is not the same as a concentration, so the caption should say "Mass mixing ratio" and not a "Concentration in ppbm" !

This has been changed in the Figure 2 caption and figure labels, and on page 11812.

Page 11844, figure 6: I do not understand what is plotted here as the terms used as different from what I am used to. A cross-section is not dimensionless. Is panel (a) showing an upscatter fraction (but the values appear too large)? Or an upscatter cross-section (define) per unit geometrical cross-section? Likewise I am not sure what an upscatter cross-section per unit volume is. This time, it seems the cross-section is not dimensionless as the unit is $\mu\mathrm{m}^{-1}$.

The text now reads “Panel (a) shows the upscatter cross-section divided by the geometric cross-section (a dimensionless ratio). Panel (b) shows the upscatter cross-section divided by the particle volume (units of μm-1)…”.