

The authors would like to thank the reviewers for their time to help improve this paper. Please see reviewer comments in italics followed by author responses in bold. First, we would like to note some changes that we, the authors, have made:

- **Title page:** Updated address for author Jason English
- **Section 2.1:** We have added a sentence noting that we used the Zhao and Turco nucleation scheme in the model.
- **Section 3.2:** We have noted that the broad injections are also slightly higher: “broader latitude and slightly higher altitude region” and added that it is responsible for part of the burden improvement: “While part of the increase in burden is due to the slightly higher injection altitude,..”
- **Section 3.3:** We have corrected “that square root” to be “the square root”.
- **Section 5:** We have corrected “reactions rates” to be “reaction rates”.

Reviewer #1: Anonymous

This is an interesting paper, presenting fundamentally new results involving the modeling of the microphysical evolution of sulfate particles for the purpose of geoengineering. However, there are a number of minor issues that have to be addressed, and they are all indicated in the comments on the attached annotated manuscript, including in the figures. After these issues are addressed, the paper should be accepted.

We have responded to each of reviewer #1’s suggestions here. (Refer to the provided page/line numbers in the reviewer highlighted changes document for his or her comments):

P2518, line 19: changed text to “rather than attempt to cool the planet through”

P 2519, line 5: no change made, since the grammatical rule is “if only part of a sentence is in quotation marks, the final quotation mark precedes the period”.

P 2519, line 11: Added Honisch et al citation (and reference).

P2520, line 5: We’ve removed the duplicate acronym definition of GCM.

P2520, line 12: We’ve corrected the spelling of Graf.

P2520, line 24: We’ve changed to “24-h” which is ACP’s desired syntax.

P2521, line 24: We’ve defined the SAGE acronym, and added the Considine et al reference.

P 2522, line 13: We include sedimentation in our model but not dry deposition. Textor et al (2006) found wet deposition was responsible for about 90% of the sulfate sink. We have added: “Prior work has found wet deposition to be

responsible for about 90% of the sulfate sink in troposphere (Textor et al., 2006); however, the absence of dry deposition in our model may impact sulfate concentrations in the boundary layer.” And in section 4 we have added: “While wet deposition is the primary tropospheric sink of sulfate aerosols (Textor et al., 2006), the lack of dry deposition in our model may introduce some error in surface perturbations.”

P 2523, line 10: We have added the sentence “Stratospheric steady-state aerosol burdens were achieved by the second simulation year”. [In the stratosphere, year-to-year variations of ambient aerosol burdens are small compared to the perturbations from geoengineering. Additionally, since injections take place directly in the stratosphere, there is not a time lag to travel from the surface to the stratosphere. For our 10 Tg S narrow SO₂ injection, steady state aerosol burden was achieved after 1 year; stratospheric burdens (Tg S) are calculated to be: 3.0 Tg (yr 1), 5.2 Tg (yr 2), 5.1 Tg (yr 3), 5.1 Tg (yr 4), 5.3 Tg (yr 5).]

P2523, line 11: This was a typo, and corrected to the 1-year period after the simulated June 14-15 eruption.

P2524, line 12: Radiative forcing is a rather straightforward calculation as a function of burden and size. As our model computes detailed microphysical aerosol properties, we report results from this perspective. We have added: “...and radiative forcing is further reduced due to a decrease of mass extinction efficiency (Heckendorn et al., 2009).”

P2524, line 23: We have added: “We weighted the effective radius by dividing the aerosol surface area in each grid box by the total vertically integrated surface area to normalize by the amount of aerosol in each grid box. Surface area was chosen since effective radius is defined by the third moment of radius by the second moment of radius.”

P2525, line 10: We have added the statement: “Indeed, our Pinatubo simulation also has higher AOD in the Northern Hemisphere, but this is not supported by observations that show a more symmetrical AOD (Minnis et al., 1993).” We have also removed the sentence from the abstract regarding the hemispheric asymmetry.

P2525 comments, and Figs 8 and 9 comments:

First, we have added a statement that our model compares favorably to Pinatubo observations: “Our model is within the error bars of Pinatubo observations of peak magnitude and timing for AOD (Ansmann et al., 1996) and effective radius (Bauman et al., 2003) in the Northern Hemisphere.”

Second, we have changed the order of paragraphs so that the discussion on hemispheric asymmetry comes after the paragraph assessing Pinatubo. We have noted that Pinatubo observations do not support our model simulations of hemispheric asymmetry, but that the Cerro Hudson eruption may convolute comparisons, and have added a reference to the Stenchikov et al paper: “Indeed,

our Pinatubo simulation also has higher AOD in the Northern Hemisphere than the Southern Hemisphere, but this is not supported by observations that show a more symmetrical AOD (Minnis et al., 1993, Stenchikov et al., 1998). In addition to the lack of QBO in our model, our model does not include the 1991 Cerro Hudson eruption in Chile, which was found to contribute to higher AOD in the Southern Hemisphere (Pitts and Thomason, 1993). A more detailed analysis of our simulation of Mount Pinatubo has been completed (English et al., 2012, in preparation).”

P2525, line 24: We have added: “This annual average was compared to the annual average of year 5 of the geoengineering simulations.”

P2526, line 12: We’ve obtained comparisons to other models and added the sentence: “For the 5 Tg injection, our model predicts effective radius in the center of the sulfate layer (50 hPa at the equator) to be 0.47 microns, compared to 0.6 microns for Heckendorn et al. (2009), and 0.4 microns for Neiemeier et al. (2010).”

P2528, line 8 etc: We have removed ~8 instances of “note” and “noted” in the text.

P2528, line 13: We have corrected the grammar from “are” to “is”.

P2528, line 26: We refer to “SO₂(4-) particles” as a shorthand notation for “hydrated sulfuric acid droplets”. We have clarified this in Table 1.

P2529, line 19: It is true that a tanker might carry over 100 tons, but these cannot reach the altitudes being discussed here. Likewise fighters may reach these levels in some boosted climb mode. However, they cannot cruise at these levels either, but would have to dump their loads in a very small region. We have added: “assuming 1 ton of H₂SO₄ per aircraft (which is a typical payload for the handful of aircraft actually able to fly at these altitudes today),”

P2531, line 20: You are correct; we had a sign convention error in our description. We’ve changed the signs.

P2534, line 15 and 16: We’ve added the tilde to “El Niño”.

P2534, line 25: We’ve capitalized “South Pole”.

P2542: We’ve added a ** footnote to describe the Pinatubo semi-lognormal injection.

Fig 1: We’ve changed “mb” to “hPa”.

Figs 2, 3, 8, 9, 10, and 15: We’ve added to the caption that geoengineering simulations are “(average of year 5)”.

Figs 3, 4, 9, 10, and 11: We've clarified the caption by changing the sentence to: "Effective radius is a column average weighted by the aerosol surface area in each grid box to apply a fair weighting to grid boxes with more surface area."

Reviewer #2: Jeffrey Pierce

This paper presents a review of how stratospheric and tropospheric sulfur burdens and optical depths will change due to stratospheric sulfur geoengineering. The authors test the sensitivity of their results to injection location and the emitted species. The paper is of interest to the ACP readership and may be published once some comments and concerns have been addressed.

P2520 L23: The global portion of Pierce et al. (2010) as well as the aerosol microphysics in Heckendorn et al. (2009) was done in a 2-D CTM.

We have corrected the text to state "2-d".

P2521 L5: "tropospheric SO₂ injections", probably more clear to call this "tropospheric anthropogenic SO₂ emissions" since "injections" to me seems more like its intentional (e.g. Geoengineering).

We have changed the text to "tropospheric anthropogenic SO₂ emissions"

P2522 L21: If you are using Euler stepping for nucleation and condensation, [H₂SO₄] would still drop by >50% within one time step even after cutting the timestep in half (and preventing [H₂SO₄] from going negative. This would cause an overprediction of the nucleation rate since the large oscillations between chemical production (increasing [H₂SO₄]) and nuc/cond would cause [H₂SO₄] to always be high at the start of the timestep. This might not matter considering the uncertainties in nucleation parameterizations and the strong dampening of particle concentrations to changes in nucleation rates (e.g. English et al. 2011).

We have added: "While our numerical model is stable, we have not done numerical tests of the accuracy of this treatment of nucleation. Since nucleation rates are very sensitive to supersaturation it is difficult to accurately predict the numbers of particles formed. However, English et al. (2011) show that even order of magnitude differences in the nucleation rates make little difference to the numbers of particles larger than about 10 nm, because even at these small sizes the particle concentrations are controlled by coagulation."

*P2526 L6: I suggest rewriting the sentence for clarity, "The particle size also increases *more dramatically* at lower levels of the stratosphere (90 hPa compared to 39 hPa)...". Also are these concentrations given at STP conditions or ambient? This information is critical for comparing aerosol distributions at different pressures.*

We've added "The particle size grows even larger at the lowest levels of the stratosphere....". These are reported at ambient conditions, similar to the Heckendorn et al paper. Since the units are reported as mass per unit volume, different levels can be compared.

P2527 L26: Line 294: Please define effective radius. I'm assuming 3rd moment of radius divided by 2nd moment of radius, but please state explicitly.

We have added a definition: "...defined as the ratio of the third moment to the second moment of the aerosol size distribution,"

P2528 L17: "latitudes" should be "longitudes".
We've changed the typo to "longitudes".

*P2529 L1: Please say "that injecting H2SO4 gas *that is instantly well-mixed throughout the gridbox* does not produce...*

Done.

P2529 L22: We did test the sensitivity to expansion/mixing rate. We did, however, assume that the plume was well-mixed radially. This would be a better uncertainty to point out.

We've removed the references to expansion rate and mixing rate. The last sentence is changed to: "Other details of the plume model, such as turbulence, may be important to the particle sizes that exit the plume, and should be validated in field studies."

P2532 L21: How good is the tropospheric aerosol simulation in WACCM/CARMA?

Our model includes the latest SO₂ surface emissions, but we don't have DMS emissions, in-cloud conversion to sulfate, or other aerosol species. Some comparisons have been completed that show the model behaves reasonably in the troposphere (English et al., 2011): SO₂ and H₂SO₄ vertical profiles are within error bars of observations from the earth's surface to the stratosphere (which vary by an order of magnitude). We don't see this significantly interfering with conclusions as the majority of the upper tropospheric perturbations are due to aerosol coming from the stratosphere, and our model does treat aerosol wet deposition. We have added a caution to section 4: "Our model does not include dimethyl sulfide (DMS) emissions, which contributes about 20% of surface sulfur emissions globally (Haywood and Boucher, 2000), or in-cloud production of sulfate. Our unperturbed simulation predicts a global atmospheric sulfate burden of 0.49 Tg S which is outside the range of IPCC simulations (0.55 to 1.1 Tg S) that include DMS and in-cloud production of sulfate (Forster et al., 2007). Therefore, fractional increases of sulfate due to geoengineering in our model may be artificially high, particularly in the high latitude Southern Hemisphere where DMS emissions peak."

Please move the discussion around P2533 L14 to the beginning of the discussion on tropospheric aerosols

We think that the discussion of DMS should remain in the paragraph discussing perturbations to high latitude Southern Hemisphere burdens, because the lack of DMS in our model would most significantly impact burdens in this region.