**Draft proposal to model emission of accumulation mode H2SO4 in**

**chemistry-climate models for a GeoMIP test-bed intercomparison study**

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**Overview**

Most modeling work exploring stratospheric aerosol injection (SAI) as a method of solar geoengineering has focused on injection of SO2. This follows from observations of Mt. Pinatubo and other eruptions demonstrating a climate response from increased sulfate aerosols. However, studies of SO2 SAI have found limitations with SO2 injection including: (1) reduced efficacy at higher loading with possible limitations on achievable RF, (2) depletion of stratospheric ozone, (3) stratospheric heating, especially in the tropical lower stratosphere, which would modify the Brewer-Dobson circulation and increase stratospheric water vapor, and (4) enhanced diffuse light at the Earth’ surface. Limitation (1) is primarily a function of the size distribution of the sulfate aerosol whereas (2)-(4) are primarily dependent on the radiative and chemical properties of sulfate though there is also some size dependence.

The size distribution problem with SO2 injection arises because condensation of H2SO4 (created by oxidation of SO2) occurs primarily onto the background aerosol particles, growing the coarse mode, increasing sedimentation, and decreasing the stratospheric lifetime of aerosols. Mean particle sizes tend to increase with the SO2 injection rate, reducing the radiative forcing efficacy (e.g., Wm-2/MtS-year-1). This problem could be reduced—and the radiative efficacy increased—if there was a way to produce additional accumulation mode particles near the optimal aerosol size distribution. One method of doing this is to inject H2SO4 vapor into a rapidly expanding aircraft plume which can lead to the formation of accumulation mode particles with a size distribution that depends on the injection rate and the expansion characteristics of the plume. Two studies, Pierce et al. [2010] and Benduhn et al. [2016] suggest—but do not prove—that appropriate size distributions can be produced in aircraft plumes using this method.

GeoMIP intercomparisons to date have focused on SO2 injection. Because SO2 injection does not represent the totality of, or necessarily the best option, for solar geoengineering, it is important that the GeoMIP community provide policy-makers with information about the potential for alternate injection methods that may reduce some of the problems with SO2 injection. To broaden the scope of GeoMIP and increase the diversity of models studying alternative injection materials and methods, we propose a geoengineering experiment with injection of H2SO4 into an aircraft wake, modeled as injection of accumulation mode sulfate aerosol particles into a GCM grid box, where the distribution of particle sizes would be consistent with values generated by models of H2SO4 injection and evolution in an expanding aircraft plume. This experiment requires minimal model development and can be performed with sectional or modal aerosol modules. In future, GeoMIP might be expanded to include stratospheric injection of solid aerosol particles, but this would require significant model development for most modeling groups and is not feasible as an intercomparison project at present.

**Two modeling challenges**

The evolution of aerosol particles after injection of H2SO4 into an aircraft wake is a problem with spatial and temporal scales from milliseconds to years, and from centimeters to global scale. No model can possibly do a good job of handling the entire problem. However, the problem can be divided into two separate domains: (a) from injection to plume dispersal, and (b) from plume dispersal to global scale. Each domain has associated uncertainties, but they can be studied separately with separate modeling tools: CFD models for domain (a) and GCMs for domain (b).

*Plume modeling.* The plume modeling problem starts with the production of small particles in a plume from the point of injection and ends when the plume has expanded sufficiently that its coalescence rate roughly matches the background stratospheric coalescence rate. The plume model needs to account for nucleation and growth of aerosol particles within an aircraft plume including the initial formation of ultrafine mode particles by homogeneous nucleation of H2SO4 and the subsequent formation of accumulation mode particles by coagulation of the ultrafine mode. In an expanding aircraft plume these processes occur on timescales of milliseconds and length scales of meters. This was addressed by Pierce et al. [2010] and then by Benduhn et al. [2016]. There is rough agreement that particles of 0.1-0.15 m radius could be produced after plume processing, but this is not certain and needs deeper investigation.

Benduhn et al. (2016) suggested that simultaneous injection of SO2 and H2SO4 may be required to achieve desired injection targets and optimal plume conditions. Coincidentally, an engineering study of the feasibility of releasing H2SO4 from an aircraft (Smith, Dykema, Keith under review) concluded that it is difficult to convert more than about 70% of S to H2SO4 onboard an aircraft with the remainder released as SO2. Thus injecting both SO2 and sulfate particles together is another sensitivity to explore. A 2-D model study (Weisenstein et al., 2017) of SAI with simultaneous injection of SO2 and particles in various proportions found that, even with only 30% of the total S released as particles of the appropriate size, substantial improvement to the overall size distribution and radiative forcing was found.

*Stratospheric models*. The second part of the problem, from injection of a given size distribution of sulfate particles into the grid of a global model to implications on aerosol burden, radiative forcing, ozone, and stratospheric temperature and circulation, can be effectively analyzed by the GeoMIP models. These models would take as input the particle size distributions that are produced as outputs from an aircraft plume study as described above. These models can address the scientific question of how this injection of new accumulation mode particles changes the large-scale particle size distribution and thus the overall radiative and dynamical response to sulfate aerosol injection. We propose an intercomparison study to address uncertainties in the global aspects of this problem.

**Proposal**

1. Model setup
   1. 10 year runs with prescribed SST.
      1. SST data set CMIP5 "SST Climatology 1988T2007" and "SEA ICE Climatology 1988T2007". Same as QBOI.
      2. 2040 GHGs and ODSs
      3. Run period: 10 year spin up, then all runs for 10 years each. 10 years is minimum - longer if you feel it necessary.
   2. AM-H2SO4 Aerosol definition
      1. **Modal models** will inject into the their accumulation mode (likely near 0.1 µm radius) with standard (for each model) mode width.
      2. **Sectional models** will translate a prescribed mode radius (0.1 µm dry) and distribution width (σ=1.5) into their aerosol size resolution
   3. Injection amount
      1. 2.5 Tg(S)/yr (optional)
      2. 5 Tg(S)/yr (**baseline)**
      3. 10 Tg(S)/yr (optional)
      4. 25 Tg(S)/yr (high)
   4. Injection locations
      1. Points: 2 points at +/- 30 degrees latitude, single longitude 180 E, 20 km altitude
      2. Region: defined by 19-21 km x 30°S to 30°N and all longitudes.
   5. Total mandatory model runs
      1. Each modeling team should run SO2 and AM-H2SO4, the *points* and *region* injection locations, and the *baseline* and *high* injection amount cases for a total of 8 runs of 10 years each, plus one spinup , or total of 90 years simulation time.
   6. Optional calculations:
      1. Mixtures of SO2 and sulfate particles: 30% sulfur mass as particles, 70% as SO2

**Requested Output:**

1. Output files and frequency: Monthly mean standard chemistry and aerosol output such as mixing ratios, aerosol effective radius, surface area density, number concentration by mode or bin, SW and LW RF. Timeseries of each variable to be in separate netcdf files.

2. Diagnostic output such as nucleation, condensation, and sedimentation rates if possible.

3. Data repository: Harvard will provide a data repository and some standard analysis of aerosols and ozone chemistry.

**References**

Benduhn, F., J. Schallock, and M. G. Lawrence (2016), Early growth dynamical implications for the steerability of stratospheric solar radiation management via sulfur aerosol particles, Geophys. Res. Lett., 43, 9956–9963, doi:10.1002/2016GL070701.

Pierce, J. R., D. K. Weisenstein, P. Heckendorn, T. Peter, and D. W. Keith (2010), Efficient formation of stratospheric aerosol for climate engineering by emission of condensible vapor from aircraft, Geophys. Res. Lett., 37, L18805, doi:10.1029/2010GL043975.

Smith, J. P., J. A. Dykema, and D. W. Keith (2017) Production of sulfates on board an aircraft: Implications for the cost and feasibility of stratospheric solar geoengineering, submitted to Earth & Space Science.

Weisenstein, D.K., Z. Dai, D.W Keith (2017) Advantages of SRM by H2SO4 injection: Scalability, steerability, and efficacy of hybrid injections, presented at Gordon Conference on Radiation Management Climate Science, Newry, Maine, 23-28 July 2017.