

An Initial Evaluation of Geoengineering by Al₂O₃ Injection

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1. Introduction

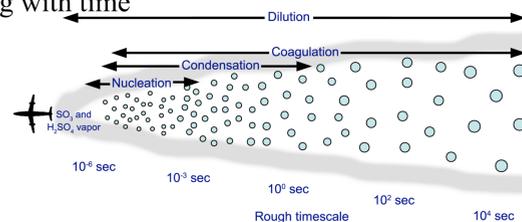
Geoengineering by injection of sulfur into the stratosphere is among the most promising methods of solar radiation management techniques under consideration [Royal Society, 2009]. However, recent studies [Heckendorn et al., 2009; Pierce et al., 2010] indicate that injections of sulfur sufficient to counteract a doubling of CO₂ would have significant adverse consequences, including tropopause heating, stratospheric water vapor increases, and ozone depletion. Alumina (Al₂O₃) particles are more efficient scatterers than sulfate particles and here are investigated for application to geoengineering. However, unlike sulfate, alumina emission has no analog in the natural stratosphere and thus may present unique and unknown risks. We provide an initial evaluation of Al₂O₃ particle evolution in a jet plume, and a sensitivity study of the impact of Al₂O₃ particles on stratospheric ozone. Even assuming that the surface area density of Al₂O₃ is smaller by factors of 5-10 than that generated by sulfate geoengineering, we find that ozone depletion is unacceptably large.

2. Modeling Approach

Expanding Plume Model for Emission into Aircraft Wake

References: Pierce and Adams [2009], Pierce et al. [2010]

- Needed for emission of condensable gases such as H₂SO₄ or alumina vapor
- Plume radially symmetric, expanding and entraining with time
- TOMAS aerosol microphysics with 43 sections: nucleation, condensation, coagulation
- All sulfur condensed within minutes, only coagulation, expansion, entrainment act after that
- Follow plume for ~2 days until coagulation with background particles becomes important
- Final size distribution depends on emission rate and plume expansion rate only



4. Al₂O₃ Aerosol Properties and Chemistry

- Efficient shortwave scattering: 4 times greater than sulfate per unit volume
- Reduced IR absorption relative to sulfate: less heating of lower stratosphere
- Al₂O₃ could be dispersed behind aircraft by condensation of combusted alumina, as occurs in solid rocket motor plumes
- Measurements show [Karasev et al., 2004] that such particles consist of micron-sized fractal aggregates of nanometer-size primary particles.
- Fractal geometry characterized by R_g (radius of gyration) and fractal dimension f_d, where aggregate mass is proportional to R_g^{f_d}
- Measured f_d=1.6 for Al₂O₃ [Karasev et al., 2004], 1.5 < f_d < 1.9 for soot [Maricq, 2006]
- Plume model of Pierce et al. [2010] modified to perform coagulation of fractal particles, assuming primary particle diameter D₀ of 10, 30, or 50 nm and fractal dimension f_d of 1.6, 2.3, or 30. Results of plume model are shown in Figure 3.

- **Heterogeneous reaction on Al₂O₃: ClONO₂ + HCl → Cl₂ + HNO₃**
 - γ = 0.02 measured by Molina et al. [1997]
 - Cl₂ rapidly dissociates in sunlight, leading to ozone depletion
- Reaction applied by Danilin et al. [2001] to solid rocket exhaust plumes
 - Calculated 0.0028% global ozone depletion with 1120 tons annual emission
- Fractal particles will have enhanced surface area density relative to spherical sulfate particles, enhancing ozone depletion
- For initial evaluation, we adopt an Al₂O₃ surface area density equivalent to that of 1 MT of SO₂ injected between 30°S-30°N, 20-25 km to represent an equivalent radiative forcing of 5 MT SO₂ injection. Results shown in Figure 4.

5. Sensitivity of Ozone to Al₂O₃ Geoengineering

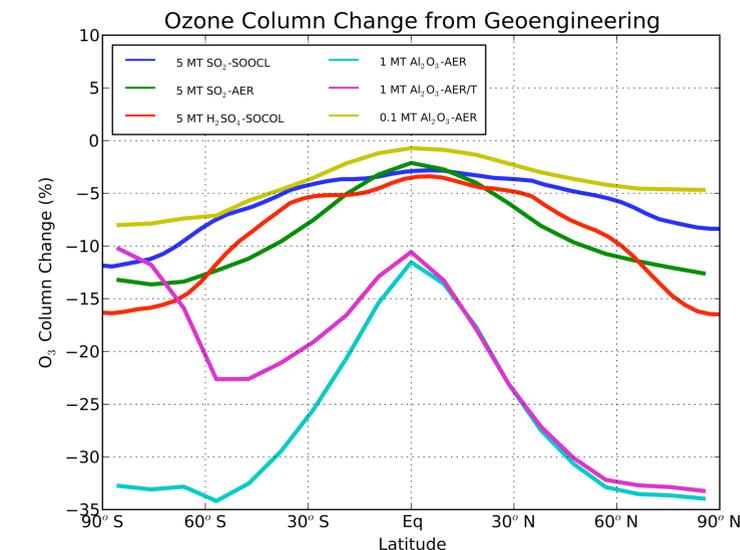
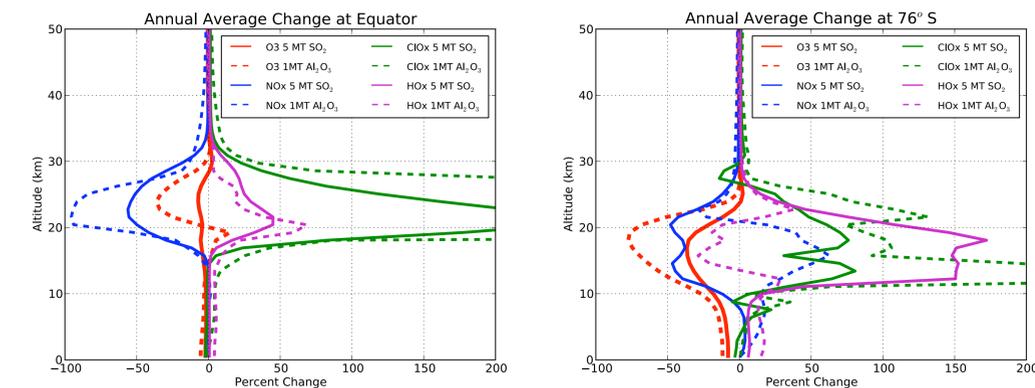


Figure 4. Calculated annual average ozone column changes in percent for several geoengineering cases. Ozone changes calculated by the SOCOL chemistry-climate model are due to changes in aerosol surface area density, changes in stratospheric temperature and circulation, and changes in water vapor and HO_x concentrations. Heckendorn et al. [2009] determined that 75% of this ozone loss is due to intensified heterogeneous chemistry caused by the increased aerosol surface area density. Ozone changes calculated by the AER chemistry-transport model are due to changes in aerosol surface area density only. Cases labeled "AER/T" used the temperature field calculated by the SOCOL model which is about 2° colder in the southern polar region. Figures below show changes in annual average O₃, NO_x, ClO_x, and HO_x concentrations at the equator and 76°S.



Global 2-D Sulfate Aerosol Model of AER

References: Weisenstein et al. [1997, 2007]

- 40 sectional aerosol size bins, 9.5 degree x 1.2 km spatial resolution
- Microphysics includes homogeneous nucleation, condensation, coagulation, evaporation, sedimentation
- Sulfur chemistry includes aerosol-gas recycling above 35 km
- Geoengineering input as either SO₂ gas or as particles defined by plume model
- Geoengineering emissions continuous in time at 30°S-30°N, 20-25 km

Ozone Change Calculations

- *SOCOL 3-D Chemistry-Climate Model* at ETH used for SO₂ and H₂SO₄ geoengineering
MA-ECHAM4 dynamics and radiation, combined with MEZON chemistry-transport
Ozone changes are due to chemistry, transport, and temperature changes
References: Schraner et al. [2008], Heckendorn et al. [2009]
- *AER 2-D Chemistry-Transport Model* used for Al₂O₃ initial evaluation
fixed circulation and temperature, full chemistry
References: Rinsland et al. [2003], Weisenstein et al. [2004]

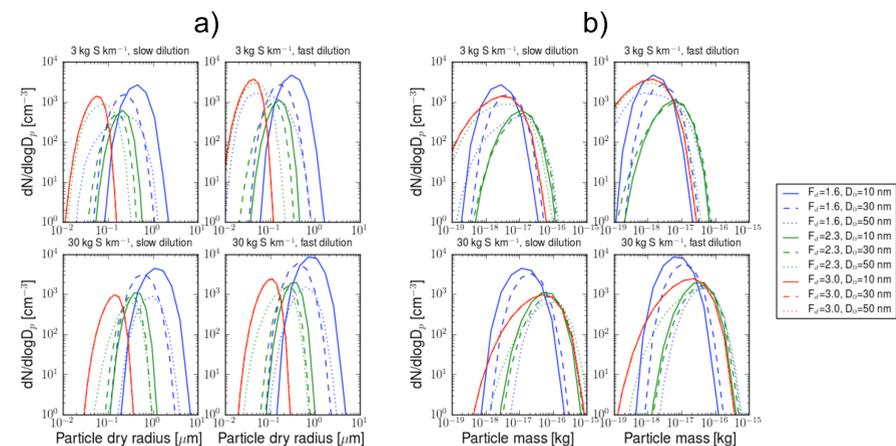


Figure 3. Calculated (a) diameter of gyration and (b) mass distribution of alumina particles 1 day after alumina vapor is sprayed from an aircraft in the stratosphere. The four panels in each figure represent uncertainties in the plume dilution rate and alumina spray rate. The various lines in each panel represent uncertainties in the fractal dimension and the size of the primary alumina particles forming the fractals. The model used for these simulations is the TOMAS microphysics model coupled to an expanding Lagrangian box model [Pierce et al., 2010].

3. Sulfate Geoengineering Results: Burdens, Flux Changes, and Surface Area Density

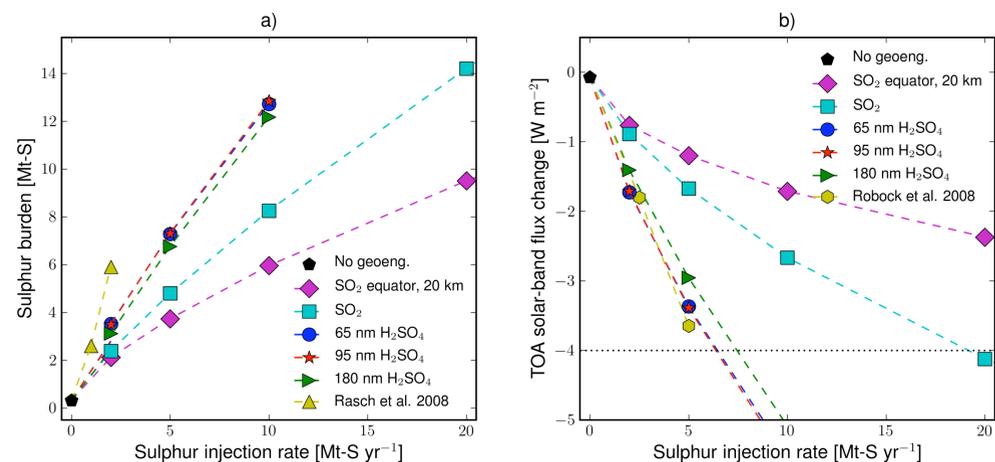


Figure 1. Calculated global annual mean (a) sulfate aerosol burdens, and (b) top-of-atmosphere solar-band flux changes as a function of geoengineering annual emission rate of sulfur, spread from 30°S-30°N, 20-25km. H₂SO₄ emissions are characterized by mode radius after 2 days of plume expansion. SO₂ emission scenarios lead to continuous nucleation and condensation, subsequently growing particles to larger sizes by coagulation and condensation onto ambient particles and yielding fast sedimentation rates and short residence times. Horizontal dashed line in panel (b) represents flux change necessary to counteract a doubling of CO₂.

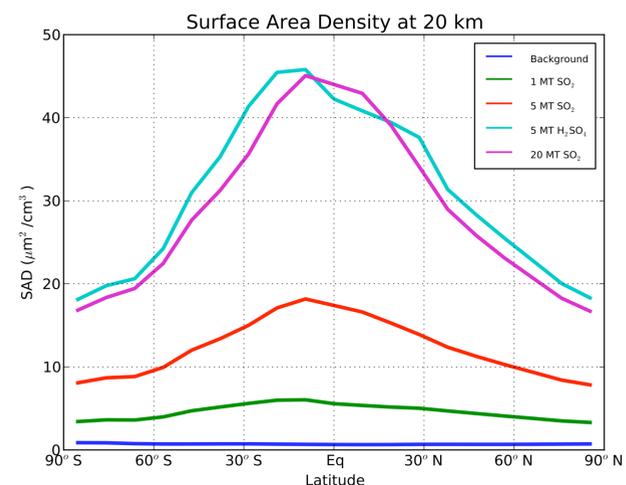


Figure 2. Calculated annual average sulfate aerosol surface area density at 20 km due to geoengineering emissions of SO₂ or of H₂SO₄ with particle mode radius of 95 nm. Emissions of 1, 5, or 20 MT/yr occur continuously in time between 30°S and 30°N and at 20-25 km. H₂SO₄ emissions into an aircraft plume produce much smaller particles than injection of SO₂, hence the greater surface area for equivalent emission rate. The radiative forcing due to a doubling of CO₂ can be approximately offset by either 7 MT/yr of H₂SO₄ emissions or 20 MT/yr of SO₂ emissions. For our sensitivity studies of ozone change due to Al₂O₃ geoengineering, we employ the surface area density of 1 MT/yr of SO₂ emissions.

6. Conclusions

- **Geoengineering is at best a partial solution** to climate change, with many unintended adverse impacts
- **Geoengineering by SO₂ injection** is very inefficient due to particle growth and sedimentation rates
- **Geoengineering by H₂SO₄ injection** into an aircraft wake can better control particle size and make -4 W/m² shortwave flux change achievable
- **Geoengineering by Al₂O₃ injection** appears to produce unacceptably high ozone loss, while ozone loss by SO₂ and H₂SO₄ geoengineering is also substantial
- **Impacts not explored** here include regional changes in surface temperature and precipitation patterns. Ocean acidification would not be ameliorated by stratospheric aerosol geoengineering.
- Adverse effects of geoengineering should be compared with the climate change resulting from **insufficient CO₂ reductions**
- **Geoengineering research** can lead to better understanding of climate change, human impacts, and volcano-climate interactions

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