An Overview of Geoengineering of Climate using Stratospheric Sulfate Aerosols

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Abstract

We provide an overview of geoengineering by stratospheric sulfate aerosols. We review the state of understanding about this topic as of early 2008, summarizing the past 30 years of work in the area, and highlight some very recent studies using general circulation models of the atmosphere and ocean, and discuss the efficacy of producing such aerosols by methods used to deliver sulfur species to the stratosphere.

The studies reviewed here all suggest that sulfate aerosols can counteract the globally averaged temperature increase associated with increasing greenhouse gases, and reduce changes to some other components of the earth system. There are likely to be remaining regional climate changes after geoengineering, with some regions experiencing significant changes in temperature or precipitation. The aerosols also serve as surfaces for heterogeneous chemistry resulting in increased ozone depletion for several decades. We conclude by highlighting many of the areas where more research is needed.

1 Introduction

The concept of “geoengineering”(the deliberate change of the Earths’ Climate by mankind (Keith, 2000)) has been considered at least as far back as the 1830s with J.P. Espy’s suggestion (Fleming, 1990) of lighting huge fires that would stimulate convective updrafts and change rain intensity and frequency of occurence. Geoengineering has been considered for many reasons since then ranging from making polar latitudes habitable to changing precipitation patterns. The history of geoengineering is reviewed elsewhere in this volume.

There is increasing concern by scientists and society in general that energy system transformation is proceeding too slowly to avoid the risk of dangerous climate change from humankind’s release of radiatively important atmospheric constituents (particularly CO_2). The assessment by the Intergovernmental Panel on Climate Change (IPCC, 2007c) shows that unambiguous indicators of human-induced climate change are increasingly evident, and there has been little societal response to the scientific consensus that reductions must take place soon to avoid large and undesirable impacts.

The first response of society to this evidence ought to be to reduce greenhouse gas emissions, but if one accepts the evidence, and notes the inertia to changing our energy infrastructure, a second step might be to explore strategies to mitigate some of the planetary warming. For this reason geoengineering for the purpose of cooling the planet is receiving increasing attention. A broad overview to geoengineering can be found in the reviews of Keith (2000), WRMSR (2007), and the papers in this volume. The geoengineering paradigm is not without its own perils (Robock, 2008). Some of the uncertainties and consequences of the approach explored here are discussed in this article. Others can be found elsewhere in this volume.

This study describes an approach to cooling the planet that goes back at least as far as 1974, when Budyko, in a series of studies (e.g., Budyko, 1974)) suggested that if global warming ever became a serious threat, we could counter it with airplane flights in the stratosphere, burning sulfur to make aerosols that would reflect sunlight away. The aerosols would increase the planetary albedo, and cool the planet, ameliorating some (but as discussed below, not all) of the effects of increasing CO_2 concentrations.

Sulfate aerosols are always found in the stratosphere. Low background concentrations arise due to transport from the troposphere of natural and anthropogenic sulfur-bearing compounds. Occasionally much higher concentrations arise from the volcanic eruptions, resulting in a temporary cooling of the Earth system (Robock, 2000), which disappears as the aerosol is flushed from the atmosphere. The volcanic injection of sulfate aerosol thus serves as a natural analog to the geoengineering aerosol. The analogy is not perfect, because the volcanic aerosol is flushed within a few years, and the climate system does not respond the same way as it would if the particles were continually replenished, as they would be in a geoengineering effort. Perturbations to the system, which might become evident with constant forcing, disappear as the forcing disappears.

This study reviews the state of understanding about geoengineering by sulfate aerosols, as of early 2008. We review the published literature, introduce some new material, and summarize some very recent results that are presented in detail in submitted articles at the time of the writing of this article. In our summary we also try to identify areas where more research is needed.

2 Review

Since the paper by Budyko (1974), the ideas generated there have received occasional attention in discussions about geoengineering (e.g., NAS92, 1992; Turco, 1995; Govindasamy and Caldeira, 2000; Govindasamy et al., 2002; Govindasamy and Caldeira, 2003; Crutzen, 2006; Wigley, 2006; Matthews and Caldeira, 2007).

There are also legal, moral, ethical, financial, and international political issues associated with a manipulation of our environment. Commentaries (Lawrence, 2006; Bengtsson, 2006; Kiehl, 2006; Cicerone, 2006; MacCracken, 2006) to Crutzen (2006) address some of these issues and remind us that this approach does not treat all the consequences of higher CO_2 concentrations (such as ocean acidification; others are discussed in Robock (2008)).

Recently, climate modellers have begun efforts to provide more quantitative assessments of the complexities of geoengineering by sulfate aerosols and the consequences to the climate system (Rasch et al., 2008; Robock et al., 2008; Tilmes et al., 2008b,a).

3 An overview of Stratospheric aerosols in the Earth System

3.1 General considerations:

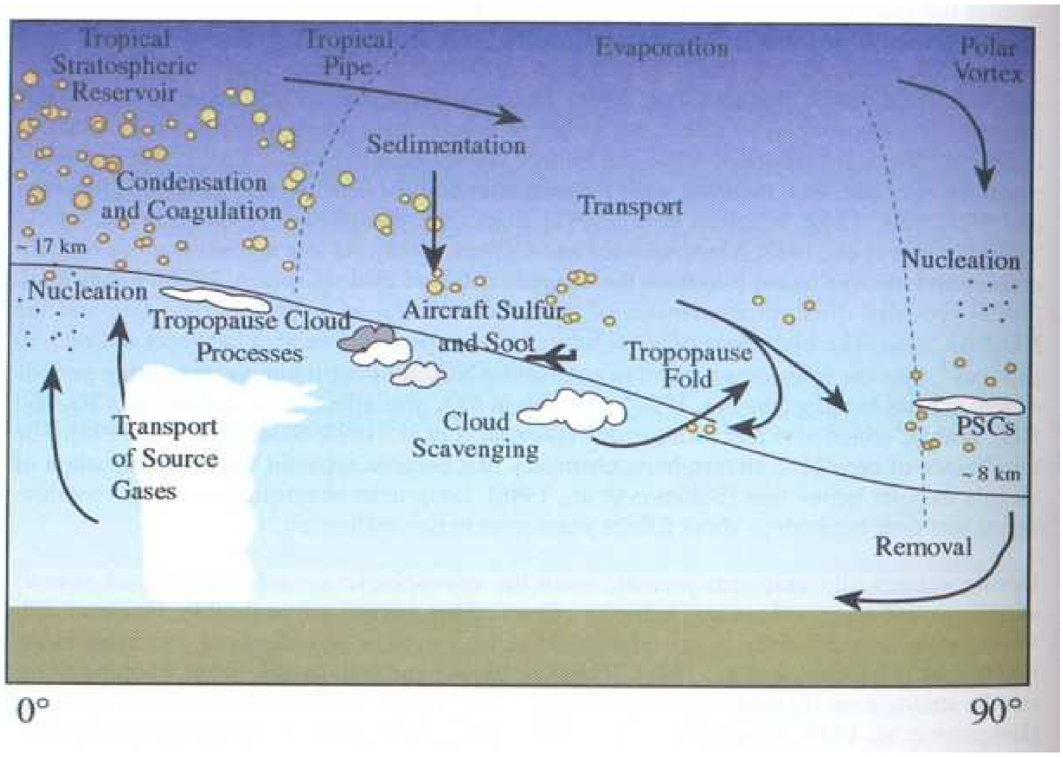

Figure 1: A schematic of the processes that influence the life cycle of stratospheric aerosols (from SPARC (2006), with permission)

Sulfate aerosols are an important component of the Earth system in the troposphere and stratosphere. Because sulfate aerosols play a critical role in the chemistry of the lower stratosphere and occasionally, following a volcanic eruption, in the radiative budget of the Earth by reducing the incoming solar energy reaching the Earth surface, they have been studied for many years. A comprehensive discussion of the processes that govern the stratospheric sulfur cycle can be found in the recent assessment of stratosphere aerosols (SPARC, 2006). Figure 1 taken from that report indicates some of the processes that are important in that region.

Sulfate aerosols play addional roles in the troposphere (IPCC, 2007c, and the references therein). As in the stratosphere they act to reflect incoming solar energy (the "aerosol direct effect"), but also act as cloud condensation nuclei, influencing the size of cloud droplets, and the persistence or lifetime of clouds (the "aerosol indirect effect"), and thus the reflectivity of clouds.

Although our focus is on stratospheric aerosols, one cannot ignore the troposphere, and so we include a brief discussion of some aspects of the tropospheric sulfur cycle also. A very rough budget describing the sources,

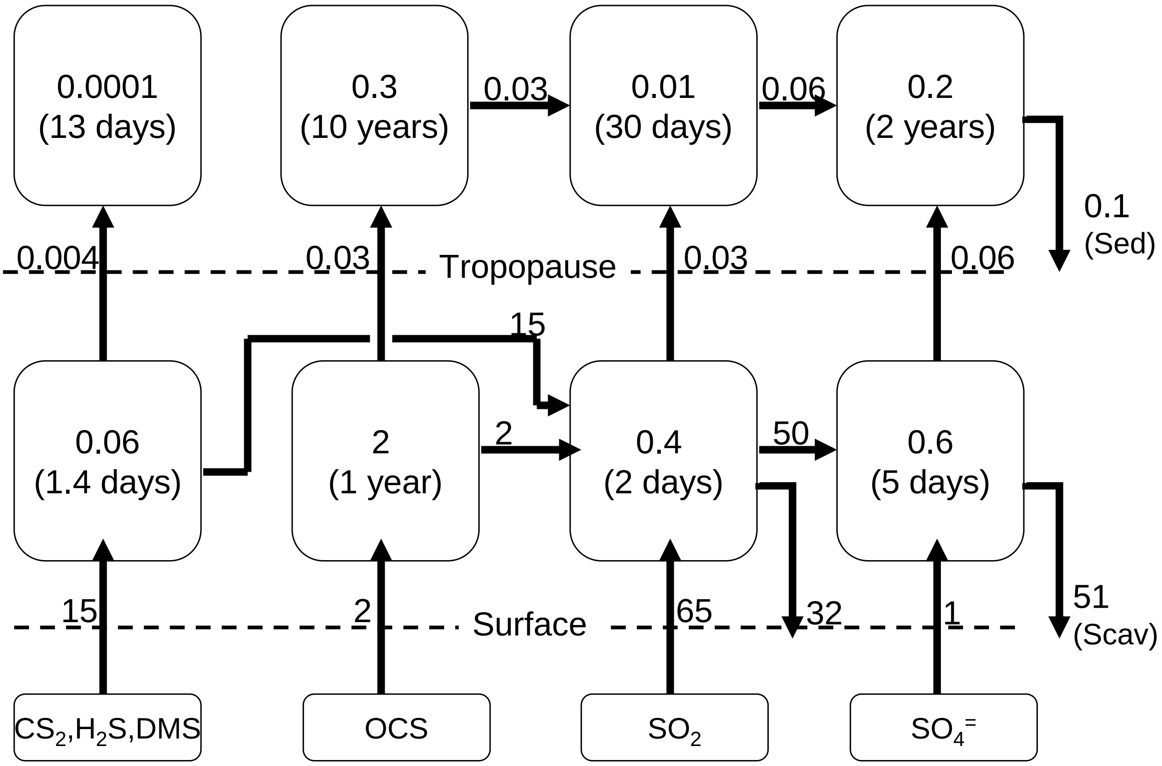

Figure 2: A very rough budget (about 1 digit of accuracy) for most of the major atmospheric sulfur species during volcanically quiescent situations, following Rasch et al. (2000),Montzka et al. (2007) and SPARC (2006). Numbers inside boxes indicate species burden in units of Tg S, and approximate lifetime against the strongest source or sink. Numbers beside arrows indicate net source or sinks (transformation, transport, emissions, and deposition processes) in Tg S/yr.

sinks, and transformation pathways during volcanically quiescent times is displayed in figure 2[1] Sources, sinks, and burdens for sulfur species are much larger in the troposphere than the stratosphere. The source of the aerosol precursors are natural and anthropogenic sulfur bearing reduced gases (DMS, SO_2, H_2S, OCS). These precursor gases are gradually oxidized (through both gaseous and aqueous reactions) to end products involving the sulfate anion (SO_4^{2-}) in combination with various other cations. In the troposphere where there is sufficient ammonia, much of the aerosol exists in the form of mixtures of ammonium sulfate ($(NH_4)_2SO_4$) and bisulfate ($(NH_4)HSO_4$).

The stratospheric sulfur bearing gases oxidize (primarily via reactions with the OH radical) to SO_2, which is then further oxidized to gaseous H_2SO_4. Stratospheric sulfate aerosols exist in the form of mixtures of condensed sulfuric acid (H_2SO_4), water, and under some circumstances, hydrates with nitric acid (HNO_3).

Although the OCS source is relatively small compared to other species, due to its relative stability, it is the dominant sulfur bearing species in the atmosphere. Oxidation of OCS is a relatively small contributor to the radiatively active sulfate aerosol in the troposphere, but plays a larger role in the stratosphere where it contributes

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[1] Sulfur emissions and burdens are frequently expressed in differing units. They are sometimes specified with respect to their molecular weight. Other time they are specified according to the equivalent weight of sulfur. They may be readily converted by multiplying by the ratio of molecular weights of the species of interest. We use only units of S in this paper, and have converted all references in other papers to these units. Also, in the stratosphere, we have assumed that the sulfate binds with water in a ratio of 75/25 H_2SO_4/water to form particles. Hence

$$3 \text{ Tg } SO_4^{2-} = 2 \text{ Tg } SO_2 = 1 \text{ Tg S} \approx 4 \text{ Tg aerosol particles.}$$

perhaps half the sulfur during volcanically quiescent conditions. Some sulfur also enters the stratosphere as SO_2, and as sulfate aerosols particles. The reduced sulfur species oxidise there and form sulfuric acid gas. The H_2SO_4 vapor partial pressure in the stratosphere – almost always determined by photochemical reactions – is generally supersaturated, and typically highly supersaturated, over its binary H_2O-H_2SO_4 solution droplets. The particles form, and grow through vapor deposition, depending on the ambient temperature, and concentrations of H_2O and H_2SO_4. These aerosol particles then are transported by winds (as are their precursors). Above the lower stratosphere, the particles can evaporate, and in the gaseous form the sulfuric acid can be photolyzed to SO_2 where it can be transported as a gas, and may again oxidize and condense in some other part of the stratosphere. Vapor deposition is the main growth mechanism in the ambient stratosphere, and in volcanic clouds, over time.

Because sources and sinks of aerosols are so much stronger in the troposphere, the lifetime of sulfate aerosol particles in the troposphere is a few days while that of stratospheric aerosol a year or so. This explains the relatively smooth spatial distribution of sulfate aerosol and resultant aerosol forcing in the stratosphere, and much smaller spatial scales associated with tropospheric aerosol.

The net source of sulfur to the stratosphere is believed to be order 0.1 Tg S/yr[1] during volcanically quiesent conditions. A volcanic eruption completely alters the balance of terms in the stratosphere. For example, the eruption of Mount Pinatubo is believed to have injected approximately 10 Tg S (in the form of SO_2) over a few days. This injection amount provides a source approximately 100 times that of all other sources over the year. The partial pressure of sulfuric acid gas consequently reaches much higher levels than during background conditions. After an eruption new particles are nucleated only in the densest parts of eruption clouds. These rapidly coagulate and disperse to concentration levels that do not aggregate significantly. Particle aggregation is controlled by Browninan coagulation (except perhaps under very high sulfur loadings). Coagulation mainly limits the number of particles, rather than the overall size of the particles, which depends more on the sulfur source strength (although considering the overall sulfur mass balance, the two processes both contribute). The particles growth is thus influenced both by vapor deposition, and proximity to other particles.

The primary loss mechanism for sulfur species from the stratosphere is believed to be sedimentation of the aerosol particles. Particle sedimentation is governed by the equations developed by Stokes in the stratosphere but requires corrections to compensate for the fact that at higher altitudes the mean free path between air molecules can far exceed the particle size, and particles fall more rapidly than they would otherwise. The aerosol particles settle out (larger particles settle faster), gradually entering the troposphere, where they are lost via wet and dry deposition processes.

Examples of the nonlinear relationships between SO_2 mass injection, particle size, and visible optical depth as a function of time assuming idealized dispersion can be found in Pinto et al. (1998). These are detailed microphysical simulations, although in a one-dimensional model with specified dispersion. The rate of dilution of injected SO_2 is critical because of the highly nonlinear response of particle growth and sedimentation rates within expanding plumes; particles only have to be 10 microns or less to fall rapidly, which greatly restricts the total suspended mass, optical depth, and infrared effect. The mass limitation indicates that 10 times the mass injection (of say Pinatubo) might result in only a modestly larger visible optical depth after some months.

The life cycle of these particles is thus controlled by a complex interplay between meteorological fields (like wind, humidity and temperature), the local concentrations of the gaseous sulfur species, the concentration of the particles themselves, and the size distribution of the particles.

In the volcanically quiescent conditions (often called background conditions) partial pressures of sulfur gases remain relatively low, and the particles are found to be quite small (Bauman et al., 2003), with a typical size distribution that can be described with a log-normal distribution with a dry mode radius, standard deviation, and effective radius of 0.05/2.03/0.17μm respectively. After volcanic eruptions when sulfur species concentrations get much higher, the particles grow much larger (Stenchikov et al., 1998). Rasch et al. (2008) used numbers for a size distribution 6-12 months after an eruption for the large volcanic-like distribution of 0.376/1.25/0.43μm following (Stenchikov et al., 1998; Collins et al., 2004), there is uncertainty in the estimates of these size distributions, and volcanic aerosol standard distribution σ_{LN} were estimated to range from 1.3 to $>$2 in Steele and Turco (1997).

When the particles are small they primarily scatter in the solar part of the energy spectrum, and play no role in heating the infrared (longwave) part of the energy spectrum. Larger particles seen after an eruption scatter and absorb in the solar wavelengths, but also absorb in the infrared (Stenchikov et al., 1998). Thus small particles tend to scatter solar energy back to space. Large particles scatter less efficiently, and also trap some of the outgoing energy in the infrared. The size of the aerosol thus has a strong influence on the climate.

3.2 Geoengineering considerations

To increase the mass and number of sulfate aerosols in the stratosphere a new source must be introduced. Using Pinatubo as an analogue, Crutzen (2006), estimated a source of 1-2 Tg S/yr would be sufficient to balance the warming associated with a doubling of CO_2. Wigley (2006) used an energy balance model to conclude that $\sim$5 Tg S/yr in combination with emission mitigation would suffice. These studies assumed that the long term response of the climate system to a more gradual injection would respond similarly to the transient response to a Pinatubo-like transient injection. A more realistic exploration can be made in a climate system model (see section 3.4).

Rasch et al. (2008) used a coupled climate system model to show that the amount of aerosol required to balance the warming is sensitive to particle size, and that nonlinearities in the climate system mattered. Their model suggested that 1.5 Tg S/yr might suffice to balance the GHG warming if the particles looked like those during background conditions (unlikely, as will be seen in section 3.3), and perhaps twice that would be required if the particles looked more like volcanic aerosols. Robock et al. (2008) used 1.5-5 Tg S/yr in a similar study, assuming larger particle sizes (which, as will be seen in the next section, is probably more realistic). They explored the consequences of injections in polar regions (where the aerosol would be more rapidly flushed from the stratosphere) and tropical injections.

All of these studies suggest that a source 15-30 times that of the current non-volcanic sources of sulfur to the stratosphere would be needed to balance warming associated with a doubling of CO_2. It is important to note that in spite of this very large perturbation to the stratospheric sulfur budget, that this is a rather small perturbation to the total sulfur budget of the atmosphere. This suggests that the deposition of the addition source of sulfur will be a very small term compared to the other sources, unless that deposition occured on a region that normally sees little deposition (perhaps the poles).

There are competing issues in identifying the best way to produce a geoengineering aerosol. Enhanced ambient aerosol can be a primary scavenger of new particles and vapors. This is a distinct disadvantage of geoengineering compared to volcanic injections, where the stratosphere is clean, the H_2SO_4 supersaturation can build up, and nucleation of new particles over time occurs more easily, with less scavenging of the new particles. Thus, the engineered layer itself becomes a limiting factor in the ongoing production of optically efficient aerosols.

Many of the earlier papers on geoengineering with stratospheric aerosols have considered delivery systems which release sulfur in very concentrated regions, using artillery shells, high flying jets, balloons, etc. These will release the sulfur in relatively small volumes of air. Partial pressures of sulfuric acid gas will get quite high, with consequences to particle growth and lifetime of the aerosols (see section 3.3 for more detail).

A third alternative would be to use a precursor gas that is quite long-lived in the troposphere but oxidizes in the stratosphere and then allow the Earth's natural transport mechanisms to deliver that gas to the stratosphere, and diffuse it prior to oxidation. OCS might serve as a natural analogue to such a gas (however it is a carcinogen).

Current sources of OCS are $\lesssim$ 1-2 Tg S/yr (Montzka et al., 2007). Perhaps 15% of that is estimated to be of anthropogenic origin. Only about $\sim$0.03-0.05 Tg S/yr is estimated to reach the tropopause and enter the stratosphere (see figure 2 and SPARC (2006)). Residence times in the troposphere are estimated to be $\sim$1-3 years, and much longer (3-10 years) in the stratosphere. Turco et al. (1980) speculated that if anthropogenic sources of OCS were to be increased by a factor of 10 that a substantial increase in sulfate aerosols would result. If we assume that lifetimes do not change (and this would require careful research in itself) then OCS concentrations would in fact need to be enhanced by a factor of 50 to produce a 1 Tg S/yr source.

It might also be possible to create a custom molecule that breaks down in the stratosphere that is not a carcinogen, but using less reactive species would produce a reservoir species that would require years to remove if society needed to stop production. Problems with this approach would be reminiscent of the climate impacts from the long lived ChloroFluoroCarbons (CFCs).

3.3 Aerosol Injection Scenarios

An issue that has been largely neglected in geoengineering proposals to modify the stratospheric aerosol is the methodology for injecting aerosols or their precursors to create the desired reflective shield.

As exemplified in section 3.4, climate simulations to date have employed specified aerosol parameters, including size, composition and distribution often with these parameters static in space and time. In this section we consider transient effects associated with possible injection schemes that utilize aircraft platforms, and estimate the microphysical and dynamical processes that are likely to occur close to the injection point in the highly concentrated injection stream. There are many interesting physical limitations to such injection schemes for vapors and aerosols, including a very high sensitivity to the induced nucleation rates (homogeneous nucleation in the case of vapor injection, which is unpredictable except very early in the injection plume).

Two injection scenarios are evaluated, both assume baseline emission equivalent to $\sim$2.5 Tg S/yr (which ultimately forms about 10 Tg of particles): 1) insertion of a primary aerosol, such as fine sulfate particles, using an injector mounted aboard an aircraft platform cruising in the lower stratosphere; and 2) sulfur-spiked fuel additives employed to emit aerosol precursors in a jet engine exhaust stream. In each case, injection is assumed to occur uniformly between 15 and 25 km, with the initial plumes distributed throughout this region to avoid hot spots. Attempts to concentrate the particles at lower altitudes, within thinner layers, or regionally — at high latitudes, for example — would tend to exacerbate problems in maintaining the engineered layer.

Our generic platform is a jet-fighter-sized aircraft carrying a payload of 10 metric tons of finely divided aerosol, or an equivalent precursor mass, to be distributed evenly over a 2500 km flight path during an four-hour flight (while few aircraft are currently capable of sustained flight at stratospheric heights, platform design issues are neglected at this point). The initial plume cross-section is taken to be 1 m^2, which is consistent with the dimensions of the platform. Note that, with these specifications, a total aerosol mass injection of 10 Tg of particles per year would call for one million flights, and would require several thousand aircraft operating continuously into the foreseeable future. To evaluate other scenarios or specifications, the results described below may be scaled to a proposed fleet or system.

Particle properties: The most optically efficient aerosol for climate modification would have sizes, R_p, on the order of 0.1 microns (μm) or somewhat less (here we will use radius rather than diameter as the measure of particle size, and assume spherical, homogeneous particles at all times). Particles this size have close to the maximum backscattering cross section per unit mass, are small enough to remain suspended in the rarified stratospheric air for at least a year, and yet are large enough and thus could be injected at low enough abundances to maintain the desired concentration of dispersed aerosol against coagulation for perhaps months (although long term coagulation and growth ultimately degrade the optical efficiency at the concentrations required — see below). As the size of the particles increases, the aerosol mass needed to maintain a fixed optical depth increases roughly as $\sim R_p$, the local mass sedimentation flux increases as $\sim R_p^4$, and the particle infrared absorptivity increases as $\sim R_p^3$. Accordingly, to achieve, and then stabilize, a specific net radiative forcing, similar to those discussed in section 3.4, larger particle sizes imply increasingly greater mass injections, which in turn accelerate particle growth, further complicating the maintenance of the engineered layer.

This discussion assumes a monodispersed aerosol. However, an evolving aerosol, or one maintained in a steady state, exhibits significant size dispersion. Upper-tropospheric and stratospheric aerosols typically have a log-normal-like size distribution with dispersion $\sigma_{LN} \sim$ 1.6–2.0 ($\ln \sigma_{LN}$ $\sim$0.47–0.69). Such distributions require a greater total particle mass per target optical depth compared to a nearly monodispersed aerosol of the same mean particle size and number concentration. Accordingly, the mass injections estimated here should be increased by a factor of $\sim$2, other things remaining equal (i.e., for σ_{LN} $\sim$1.6–2.0, the mass multiplier is in the range of 1.6–2.6).

Aerosol microphysics: A bottleneck in producing an optically efficient uniformly dispersed aerosol — assuming perfect disaggregation in the injector nozzles — results from coagulation during early plume evolution. For a delivery system with the specifications given above, for example, the initial concentration of plume particles of radius R_{po} =0.08 μm would be $\sim$1x10^9/cm^3, assuming sulfate-like particles with a density of 2 g/cm^3. This initial concentration scales inversely with the plume cross-sectional area, flight distance, particle specific density, and cube of the particle radius, and also scales directly with the mass payload. For example, if R_{po} were 0.04 μm or 0.16 μm, the initial concentration would be $\sim$1x10^{10}/cm^3 or 1x10^8/cm^3, respectively, other conditions remaining constant.

For an injected aerosol plume, the initial coagulation time constant is,

$$t_{co} = \frac{2}{n_{po} K_{co}} \tag{1}$$

where n_{po} is the initial particle concentration (#/cm^3) and K_{co} is the self-coagulation kernel (cm^3/sec) corresponding to the initial aerosol size. For R_{po} $\sim$0.1 μm, K_{co} $\sim$3x10^{-9} cm^3/sec (e.g., Turco et al., 1979; Yu and Turco, 2001). Hence, in the baseline injection scenario, t_{co} $\sim$0.07–7 sec, for R_{po} $\sim$0.04–0.16 μm, respectively. To assess the role of self-coagulation, these time scales must be compared to typical small-scale mixing rates in a stably-stratified environment, as well as the forced mixing rates in a jet exhaust wake.

Turco and Yu (1997, 1998, 1999) derived analytical solutions of the aerosol continuity equation that describe the particle microphysics in an evolving plume. The solutions account for simultaneous particle coagulation and condensational growth under the influence of turbulent mixing, and address the scavenging of plume vapors and particles by the entrained background aerosol. A key factor — in addition to the previous specifications — is the growth, or dilution, rate of a plume volume element (or, equivalently, the plume cross-sectional area). The analytical approach incorporates arbitrary mixing rates through a unique dimensionless parameter that represents the maximum total number of particles that can be maintained in an expanding, coagulating volume element at any time. Turco and Yu (1998, 1999) show that these solutions can be generalized to yield time-dependent particle size distributions, and accurately reproduce numerical simulations from a comprehensive microphysical code. Although aerosol properties (concentration, size) normally vary across the plume cross-section (e.g., Brown et al., 1996; Dürbeck and Gerz, 1996), uniform mixing is assumed, and only the mean behavior is considered.

Quiescent injection plumes: An otherwise passive (non-exhaust) injection system generally has limited turbulent energy, and mixing is controlled more decisively by local environmental conditions. If the quiescent plume is embedded within an aircraft wake, however, the turbulence created by the exhaust and wing vortices can have a major impact on near-field mixing rates (e.g., Schumann et al., 1998). For a quiescent plume, we adopt a linear cross-sectional growth model that represents small scale turbulent mixing perpendicular to the plume axis (e.g., Justus and Mani, 1979). Observations and theory lead to the following empirical representation for the plume volume,

$$V\left(t\right)/V_o = \left(1 + t/\tau_{mix}\right) \tag{2}$$

where V is the plume volume element of interest (equivalent to the cross-sectional area in the near-field), V_o is its initial volume, and τ_{mix} is the mixing time scale. For the situations of interest, we estimate 0.1$\leq$ τ_{mix} $\leq$ 10 sec.

Following Turco and Yu (1999; Eq. 73), we find for a self-coagulating primary plume aerosol,

$$N_p\left(t\right)/N_{po} = \frac{1}{1 + f_m \ln\left(1 + f_c/f_m\right)} \tag{3}$$

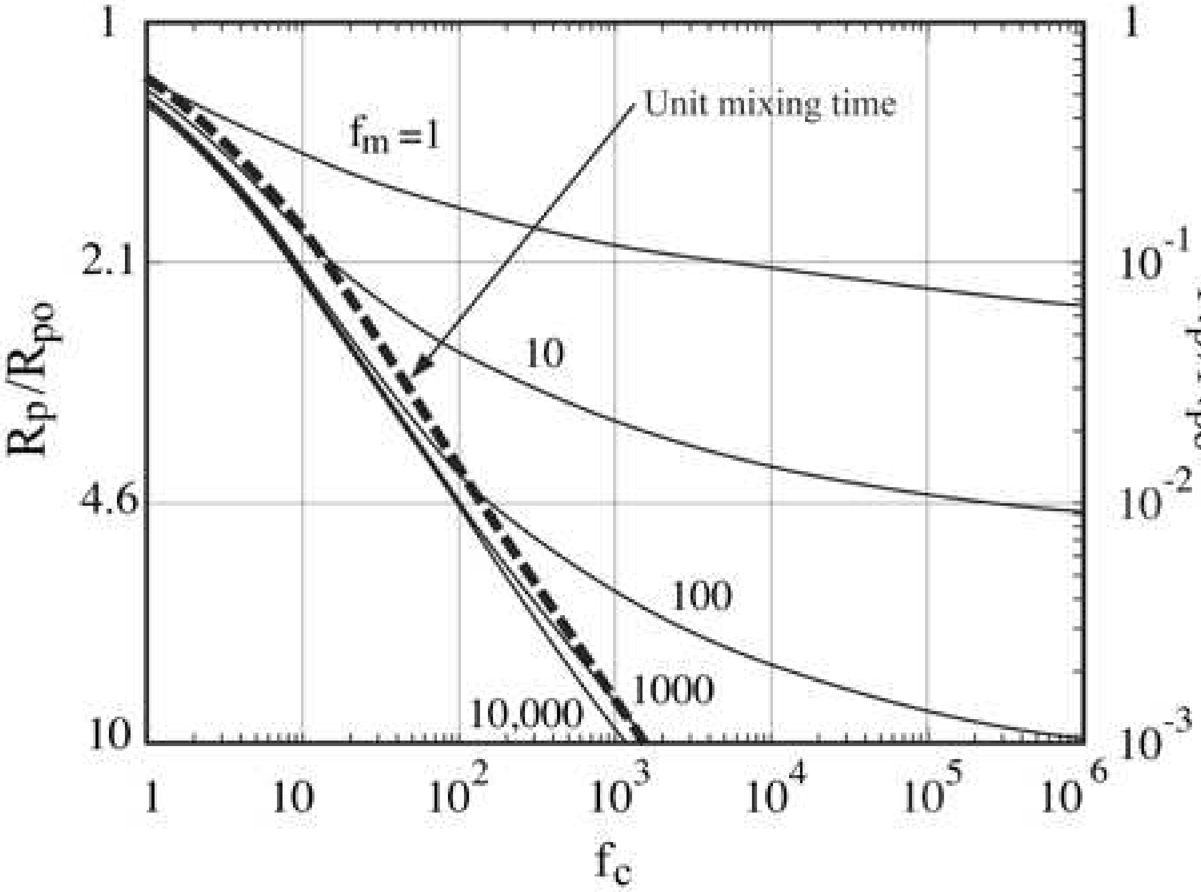

Figure 3: Evolution of the total concentration of particles N_p and the mass-mean particle radius R_p in an expanding injection plume. Both variables are scaled against their initial values in the starting plume. The time axis ($f_c = t/\tau_{co}$) is scaled in units of the coagulation time constant τ_{co}. Each solid line, corresponding to a fixed value of f_m gives the changes in N_p and R_p for a specific mixing time scale τ_{mix} measured relative to the coagulation time scale τ_{co} or $f_m = \tau_{mix}/\tau_{co}$. The heavy dashed line shows the changes at the unit mixing time, for which $f_c = f_m$ when the plume cross sectional area has roughly doubled; the longer the mixing time scale, the greater the reduction in particle abundance and particle radius.

where N_p is the total number of particles in the evolving plume volume element at time, *t*, and N_{po} is the initial number. We also define the scaled time, $f_c = t/\tau$, and scaled mixing rate, $f_m = \tau_{mix}/\tau_{co}$. The local particle concentration is, $n_p(t) = N_p(t)/V(t)$.

In Figure 3, predicted changes in particle number and size are illustrated as a function of the scaled time for a range of scaled mixing rates. The ranges of parameters introduced earlier result in an approximate range of 0.014 $\leq f_m \leq$ 140. At the lower end, prompt coagulation causes only a small reduction in the number of particles injected, while at the upper end, reductions can exceed 90% in the first few minutes. Particle self-coagulation in the plume extending over longer time scales further decreases the initial population — by a factor of a thousand after one month in the most stable situation assumed here, but only by some 10's of percent for highly energetic and turbulent initial plumes.

The dashed line in Figure 3 shows the effect of coagulation at the "unit mixing time," at which the plume volume has effectively doubled. Clearly, prompt coagulation significantly limits the number of particles that can be injected into the ambient stratosphere when stable stratification constrains early mixing. Initial particle concentrations in the range of $\sim10^{10}$–10^{11}/cm^3 would be rapidly depleted, as seen by moving down the unit mixing time line in Figure 3 (further, 10^{11}/cm^3 of 0.08 μm sulfate particles exceeds the density of stratospheric air). A consequence of prompt coagulation is that it is increasingly difficult to compensate for plume coagulation (at a fixed mass injection rate) by reducing the starting particle size. Initial particle concentrations could simultaneously be reduced to offset coagulation, but the necessary additional flight activity would impact payload and/or infrastructure.

Aerosol injection in aircraft jet exhaust. The effects of high-altitude aircraft on the upper troposphere and lower stratosphere have been extensively studied, beginning with the supersonic transport programs of the 1970's and extending to recent subsonic aircraft impact assessments (under various names) in the US and Europe (e.g. NASA-AEAP, 1997). These projects have characterized aircraft emissions and jet plume dynamics, and developed corresponding models to treat the various chemical, microphysical and dynamical processes.

Spiking aircraft fuel with added sulfur compounds (H_2S, S_n) could enhance the particle mass in a jet wake. It is well established that ultrafine sulfate particles are generated copiously in jet exhaust streams during flight (e.g. Fahey et al., 1995). The particles appear to be nucleated by sulfuric acid on chemiions generated in the engine combustors (Yu and Turco, 1997, 1998b). Sulfuric acid is a byproduct of sulfur residues in the fuel (typically $<$1% sulfur by weight); most of this fuel sulfur is emitted as SO_2. The fraction emitted as H_2SO_4 decreases as the fuel sulfur content increases, and accounts for roughly 2% of the total sulfur as the fuel sulfur content approaches $\sim$1%.

The concentrations of chemiions in jet emissions are strongly limited by ion-ion recombination along the engine train to $\sim$1x10^9/cm^3 (both $\pm$) at the exit plane (e.g., Arnold et al., 2000). Considering a variety of direct measurements of particles in jet wakes, Kärcher et al. (2000) showed that chemiion nucleation is consistent with the observed relative constancy of the ultrafine volatile (non-soot) particle emission factor, $E_p \sim$ 1-2x10^{17}/kg-fuel (where it should be noted that the concentrations of soot particles are typically $<$1% of the total number of particles emitted). E_p is quite insensitive to the fuel sulfur content, a fact that is also consistent with a chemiion nucleation source. While contrails formed in jet wakes can significantly modify the injected particle properties (e.g., Yu and Turco (1998a)), condensation trails are extremely rare under normally dry stratospheric conditions.

If we imagine enhanced jet fuel sulfur contents of 5% by weight (10-100 times current amounts) for geoengineering purposes, then the annual consumption of about 50 Tg of such fuel during stratospheric flight (about half the amount used by current commercial aviation) could emit up to 2.5 Tg of sulfur that would eventually generate roughly 10 Tg of sulfate aerosol. The total number of particles emitted — for $E_p \sim$ 1x10^{17}/kg-fuel — would amount to $\sim$5x10^{27}. This number, uniformly dispersed over a 10-km thick layer from 15–25 km, yields an average concentration of $\sim$1x10^3/cm^3 with a particle radius of roughly 0.06 μm; in other words, an ideal geoengineered solar shield. These estimates: (i) assume no unexpected chemistry or microphysics in the early wake that would alter the emission factor significantly; (ii) allow for an ideal distribution of sulfate mass among the particles; and (iii) ignore coagulation following emission.

The mixing rates in a jet wake are very rapid. Schumann et al. (1998) fit a wide range of exhaust plume observations in the upper troposphere and lower stratosphere with a "universal" mixing curve. We use their result in the form,

$$V/V_o = 100t^{0.8}; t \geq 0.0032sec \tag{4}$$

Eq. (4) describes, roughly, plume dilution starting at the exhaust exit prior to mixing with turbine bypass air, through the jet zone, vortex region, and into the ambient mixing regime. Schumann et al. (1998) state that the fit is best between 1 and 50 seconds. For the $\sim$1x10^9/cm^3 incipient particles in the initial exhaust stream, the extent of self-coagulation can be projected using the more general analytical approach discussed earlier (Turco and Yu, 1999). Thus, even at 10^5 sec, about three-quarters of the initial particles remain (compared to an estimated 0.0007% if mixing were completely suppressed). Clearly, prompt coagulation is not an issue in a jet exhaust plume.

Longer-term plume processing: The extended microphysical processing of an injection plume can be critical because of the long induction time before the plume becomes widely dispersed as part of the background aerosol. Yu and Turco (1999) studied the far-wake regime of jet exhaust for upper tropospheric conditions to estimate the yield of cloud condensation nuclei from volatile aircraft particulate emissions. In their simulations, the background aerosol surface area density ranged from 12.7–18.5 μm^2/cm^3 for summer conditions. The resulting scavenging of fresh plume particles amounted to about 95% after 10 days (that is, the effective emission index was decreased by a factor of 20). Moreover, only about 1 in 10,000 of the original particles had grown to 0.08 μm at that time, corresponding to a fuel sulfur content of 0.27% by weight, with 2% emitted as H_2SO_4. For a geoengineering scheme with 5% fuel sulfur, although the primary exhaust sulfuric acid fraction would probably be less than one percent, the initial growth rate of the chemiions would likely be accelerated.

At typical mixing rates, background aerosol concentrations would be present in an injection plume within a minute or less. The natural stratosphere has an ambient aerosol concentration of 1-10/cm^3, with an effective surface area of <1 μm^2/cm^3. However, in a geoengineered stratosphere, at the desired baseline optical depth, a surface area density >10 μm^2/cm^3 would prevail. Further, any attempt to concentrate the engineered layer regionally or vertically, or both, would greatly exacerbate both self-coagulation and local scavenging.

The coagulation kernel for collisions of the background engineered particles (assuming a minimum radius of $\sim$0.1-0.2 μm following aging) with jet exhaust nanoparticles of $\sim$10–80 nm is $\sim$1x10^{-7} – 4x10^{-9} cm^3/sec, respectively (Turco et al., 1979). Using a mean scavenging kernel for growing jet particles of $\sim$2x10^{-8} cm^3/sec, and a background concentration of 120/cm^3 (estimate for a doubling of the mass injection rate to maintiin the optical depth, see below), the estimated scavenging factor is $exp(-2.5 \times 10^{-6t})$. After one day, the reduction in number is a factor of $\sim$0.80, and over ten days, $\sim$0.1, consistent with the result of Yu and Turco (1999). Keeping in mind that the optical requirements of the engineered layer are roughly based on total cross section (ignoring infrared effects), while the scavenging collision kernel is also roughly proportional to the total background particle surface area (for the particle sizes relevant to this analysis), larger particles imply a lower concentration (and greater injection mass loading) but about the same overall scavenging efficiency.

The background aerosol will also affect the partitioning of any injected vapors between new and pre-existing particles. Considering the injection of SO_2 in jet exhaust as an example, it should be noted that SO_2 oxidation in the stabilized plume roughly a day, unless oxidants are purposely added to the plume. By this time the SO_2 would be so dilute and relative humidty so low that additional nucleation would be unlikely.

At about 1 day, the residual plume exhaust particles may have achieved sizes approaching 0.05 μm (Yu and Turco, 1999). Then, considering the considerably larger surface area of the background aerosol, only a fraction of the available precursor vapors would migrate to new particles, with the rest absorbed on pre-existing aerosol. Using an approach similar to that in Turco and Yu (1999), we infer that the jet-fuel sulfur injection scenario partitions roughly 20% of the injected sulfur onto new particles, with the rest adding to the background mass. Considering the higher fuel sulfur content, and reduced number of condensation sites, the residual injected plume particles could grow on average to about $\sim$0.08 μm. While this is a desirable size, the effective emission index is an order of magnitude below that needed to maintain the desired layer under the conditions studied. Either the fuel sulfur content or fuel consumption could be doubled to regain the overall target reflectivity. Nevertheless, as the expanding injection plumes merge and intermix following the early phase of coagulation scavenging, the aerosol system undergoes continuing self-coagulation as the layer approaches, and then maintains, a steady state. The consequences of this latter phase are not included in these estimates.

Summary: A primary conclusion of the present analysis is that the properties of aerosols injected directly into the stratosphere from a moving (or stationary) platform, or in the exhaust stream of a jet aircraft, can be severely affected by prompt and extended microphysical processing as the injection plume disperses, especially due to self-coagulation and coagulation scavenging by the background aerosol. Early coagulation can increase mass requirements because of increased particles sizes by a factor of two or more. In addition, the resulting dispersion in particle sizes implies even greater mass injections by up to $\sim$2. As a result, the extent of the engineering effort and infrastructure development needed to produce the required net solar forcing would exceed optimum levels by an overall factor of at least several, and perhaps more in non-ideal circumstances.

3.4 Global Modelling

Most of the studies mentioned in the previous sections calibrated their estimates of the climate response to geoengineering aerosol (Crutzen, 2006; Wigley, 2006) based upon historical observations of the aerosol produced by volcanic eruptions. Crutzen and Wigley focussed primarily upon the surface temperature cooling resulting from the aerosol's shielding effect. Trenberth and Dai (2007) analyzed historical data to estimate the role of the shielding on the hydrological cycle, and concluded that there would be a substantial reduction in precipitation over land, with a consequent decrease in runoff and river discharge to the ocean.

The analogy between a volcanic eruption and geoengineering via a sulfate aerosol strategy is imperfect. The aerosol forcing from an eruption lasts a few years at most, and eruptions occur only occasionally. There are many timescales within the Earth system, and their transient response to the eruption is not likely to be the same as the response to the continuous forcing required to counter the warming associated with greenhouse gases. Furthermore we have no precise information on the role the eruptions might have on a world warmer than today. For example, the response of the biosphere to a volcanic eruption might be somewhat different in a warmer world than it is today. It is thus of interest to explore the consequences of geoengineering using a tool (albeit flawed) that can simulate some of the complexities of the Earth system, and ask how the Earth's climate might change were one to successfully introduce particles into the stratosphere.

Govindasamy and Caldeira (2000); Govindasamy et al. (2002); Govindasamy and Caldeira (2003) and Matthews and Caldeira (2007) introduced this line of exploration, mimicking the impact of stratospheric aerosols by reducing the solar constant to diminish the energy entering the atmosphere (by 1.8%). These studies are discussed in more detail elsewhere in this volume so we will not review them further here.

Rasch et al. (2008) used a relatively simple representation of the stratospheric sulfur cycle to study this problem. The aerosol and precursor distributions evolution is controlled by production, transport, and loss processes as the model atmosphere evolves. The aerosols are sensitive to changes in model climate and this allows some feedbacks to be explored (for example changes in temperature of the tropical tropopause, and lower stratosphere, and changes to cross tropopause transport). Their model used a "bulk" aerosol formulation carrying only the aerosol mass (the particle size distribution was prescribed). They used a coupled Atmosphere Ocean General Circulation Model (AOGCM) variant of the NCAR Community Atmosphere Model (CAM3) (Collins et al., 2006), coupled to a slab ocean model (SOM). The model was designed to produce a reasonable climate for the troposphere and middle atmosphere. The use of a SOM with a thermodynamic sea ice model precluded a dynamic response from the ocean and sea-ice, requiring a more complex model like that of Robock et al. (2008) discussed below.

The model was used to explore the evolution of the sulfate aerosol and the climate response to different amounts of precursor injection, and the size of the aerosol. SO_2 was injected uniformly and continuously in a 2 km thick region at 25 km between 10°N and 10°S. Because of the difficulties of modelling the particle size evolution discussed in section 3.3 the study assumed the distribution to either be "small", like that seen during volcanically quiescent situations or "large" like particles seen following an eruption. Figure 4 shows the aerosol distribution and radiative forcing for an example simulation (assuming a 2Tg S/yr source and particle size similar to a volcanic aerosol). We have chosen to focus on the June, July, August season to highlight some features that disappear when displaying annual averages. The aerosol is not distributed uniformly in space and time. The mass of aerosol is concentrated in equatorial regions near the precursor injection source region, and in polar regions where the volume of air is optimal for the existance of aerosol, and away from the mid-latitude regions with relatively rapid exchange with the troposphere. Aerosol burdens are highest in the winter hemisphere, but because solar insolation is lower there, radiative forcing is also lower than in the summer hemisphere. Maximum radiative forcing occurs in the high latitudes of the summer hemisphere, acting to effectively shield the high latitudes resulting in a substantial recovery of sea ice compared to the $2xCO_2$ scenario (see Rasch et al. (2008)).

While the largest forcing in the annually averaged sense occurs in equatorial regions, the seasonal forcing is largest in the summer hemisphere, the most sensitivity in the response occurs at the poles, consistent with the general behavior of climate models to uniform radiative forcing from greenhouse gases (IPCC, 2007c), and also to the response to volcanic eruptions (Robock, 2000), and to simpler explorations of geoengineering (Govindasamy and Caldeira, 2000). Stratosphere Troposphere Exchange (STE) processes respond to greenhouse gas forcing and interacts with geoengineering. Nonlinear feedbacks modulate STE processes and influence the amount of aerosol precursor required to counteract CO_2 warming. They found that $\sim$50% more aerosol precursor must be injected than would be estimated if STE processes did not change in response to greenhouse gases or aerosols. Aerosol particle size was also found to play a role. More aerosol mass ($\sim$100%) is required to counteract greenhouse warming if the aerosol particles become as large as those seen during volcanic eruptions, because larger particles are less effective at scattering incoming energy, and trap some of the outgoing energy. 2 Tg S/yr was estimated to be more than enough to balance the warming in global-mean terms from a doubling of CO_2 if particles were small (probably unlikely), but insufficient if the particles are large. Small particles were optimal for geoengineering through radiative effects, but also provided more surface area for chemistry to occur. The reduced single scattering albedo of the larger particles and increased absorption in the infrared lessen the impact of the geoengineering, making large particle sulfate less effective in cooling the planet. That study also indicated the potential for ozone depletion. Ozone depletion issues are discussed in more detail in section 3.4.1.

A typical surface temperature change from present day for a $2xCO_2$ scenario is shown in figure 5 along with the result of geoengineering at 2 Tg S/yr (assuming a volcanic sized particle) . The familiar CO_2 warming signal, particularly at high latitude is evident, with a substantial reduction resulting from geoengineering. The simulation uses an emission rate that is not sufficient to completely counterbalance the warming. Geoengineering at this amplitude leaves the planet 0.25-0.5K warmer than present over most of the globe, with the largest warming remaining at the winter pole. It is also straightforward to produce an emission that is sufficient to overcool the model (e.g. Rasch et al. (2008)). The polar regions, and continents show the most sensitivity to the amplitude of the geoengineering.

Robock et al. (2008) (hereafter referred to as the “Rutgers” study) moved to the next level of sophistication in modeling geoengineering on the climate system. They used the GISS atmospheric model (Schmidt et al, 2006) and included a similar formulation for sulfate aerosols (Oman et al., 2005, 2006a,b) with a substantially lower horizontal (4x5 degree) and vertical (23 layers to 80km) spatial resolution than Rasch et al. (2008). Instead of using a slab ocean and sea ice model, they included a full ocean and sea ice representation. While Rasch et al. (2008) examined the steady state response of the system for present and doubled CO_2 concentrations, Robock et al. (2008) explored solutions with transient CO_2 forcings using an IPCC A1B scenario with transient greenhouse gas forcing. They examined the consequences of injections of aerosol precursors at various altitudes and latitudes to a 20 year burst of geoengineering, between 2010 and 2030. We focus on two of their injection scenarios: 1) an injection of 2.5 Tg S/yr in the tropics at altitudes between 16-23 km; 2) an injection of 1.5 Tg S/yr at latitude 68°N between 10-15 km. They chose a dry mode radius of 0.25 μm, intermediate to the ranges explored in the Rasch et al. (2008) study. The midlatitude injection produces a shorter lifetime for the aerosol, and concentrates its impact on the Arctic, although, as they show (and as seen below) it has global consequences. This type of geoengineering scenario shares some commonalities with scenarios described by Caldeira elsewhere in this volume. Robock et al. (2008) also showed that geoengineering is able to return sea-ice, surface temperature, and precipitation patterns to values closer to the present day values in a climate system model.

As an example, we show changes in precipitation for a few scenarios from Robock et al. (2008) and Rasch et al. (2008) in Figure 6, again for a JJA season. Because the signals are somewhat weaker than evident in the surface temperature changes shown above, we have hatched areas where changes exceed 2 standard deviations of an ensemble of control simulations to indicate differences that are likely to be statistically important. The top row shows results from the NCAR model from Rasch et al. (2008), the bottom (labeled Rutgers) shows results from the GISS model as described in Robock et al. (2008).

As noted in IPCC (2007a), projections of changes from forcing agents to the hydrologic cycle through climate models is difficult. Uncertainties are larger than in projections of temperature, and important deficiencies remain in the simulation of clouds, and tropical precipitation in all climate models, both regionally and globally, so results from models must be interpreted carefully and viewed cautiously. Nevertheless, climate models do provide

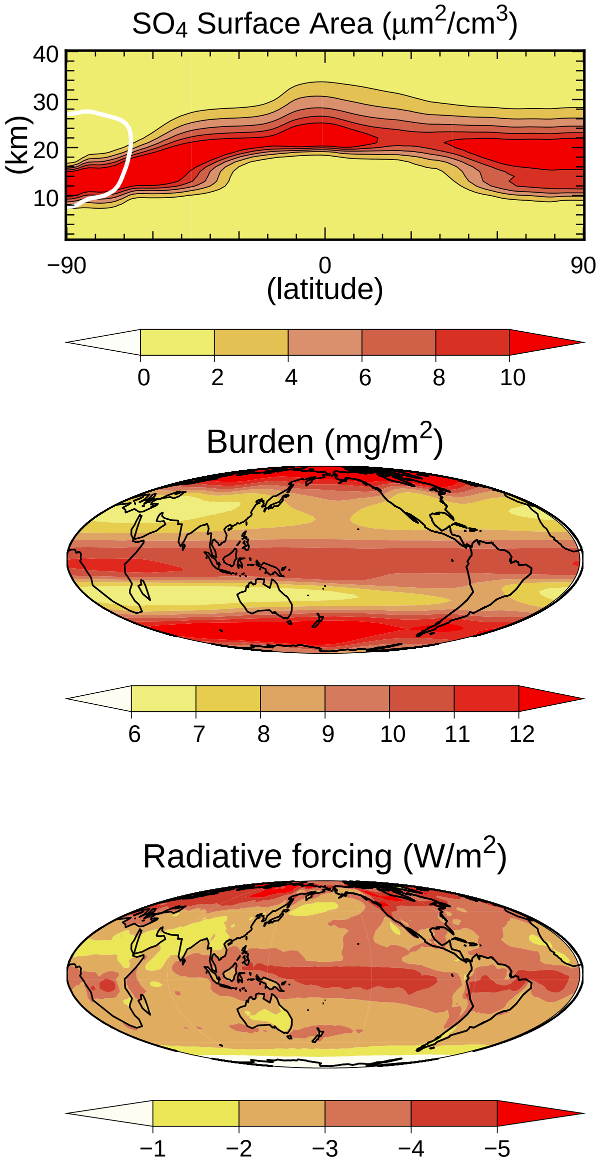

Figure 4: Examples of distribution of the geoengineering aerosol for June, July, August from a 20 year simulation for a 2 Tg S/yr emission. The white contour in the top panel shows the region where temperatures fall below 194.5 K, and indicate approximately where ozone depletion may be important (see section 3.4.1).

information about the fundamental driving forces of the hydrologic cycle and its response to changes in radiative forcing (e.g. Annamalai et al. (2007)).

The NCAR results (top left panel), consistent with IPCC (2007b) and the 20+ models summarized there, suggests a general intensification in the hydrologic cycle in a doubled CO_2 world with substantial increases in regional maxima (such as monsoon areas) and over the tropical Pacific, and decreases in the subtropics. Geoengineering (top right panel, in this case not designed to completely compensate for the CO_2 warming), reduces the impact of the warming substantially. There are many fewer hatched areas, and the white regions indicating differences of less than 0.25 mm/day are much more extensive)

The Rutgers simulations show a somewhat different spatial pattern, but again, the perturbations are much

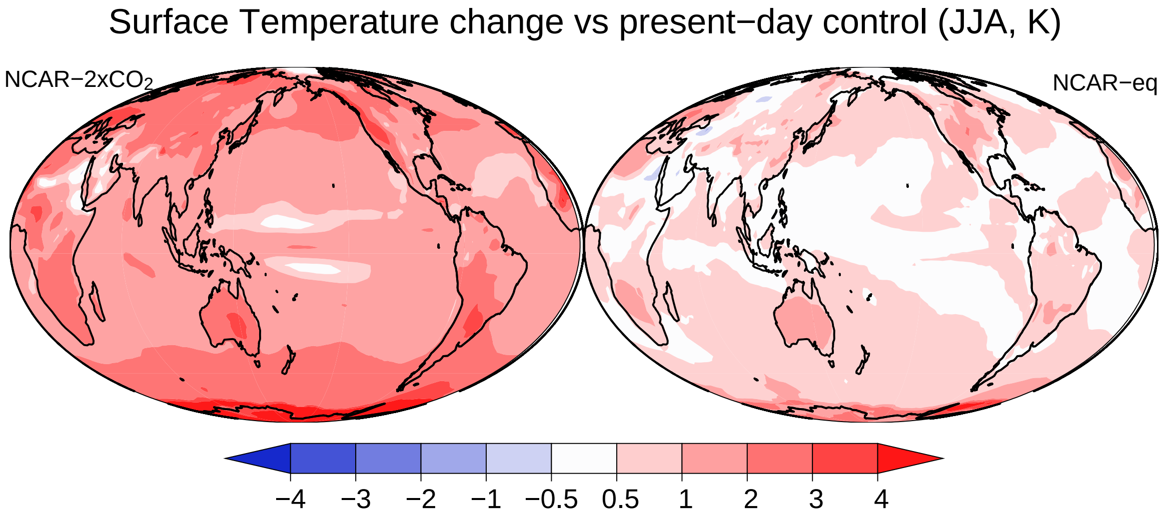

Figure 5: The surface temperature difference from present day during June, July, August the 2xCO_2 simulation and the geoengineering simulation using 2 Tg S/yr emission (which is not sufficient to entirely balance the greenhouse warming).

smaller than those evident in an "ungeoengineered world" with CO_2 warming. The lower left panel shows the precipitation distributions for the polar injection, the lower right the distributions for the equatorial injection. Both models show changes in the indian and SE asian monsoon regions, and common signals in the equatorial Atlantic. There are few common signals between the NCAR and Rutgers estimates. Robock et al. (2008) have emphasized that the perturbations that remain in the monsoon regions after geoengineering are considerable and expressed concern that these perturbations would influence the lives of billions of people. This would certainly be true. However, it is important to keep in mind: 1) that the perturbations after geoengineering are smaller than those without geoengineering; 2) the remaining perturbations are $\leq$ 0.5mm/day in an area where seasonal precipitation rates reach 6-15mm/day; 3) the signals differ between the NCAR and Rutgers simulations in these regions; and 4) monsoons are a notoriously difficult phenomenon to model (Annamalai et al., 2007). These caveats only serve to remind the reader about the importance of a careful assessment of the consequences of geoengineering, and the general uncertainties of modeling precipitation distributions in the context of climate change.

3.4.1 Impact on chemistry and the middle atmosphere

Historically, most attention has focussed on the surface chemistry responsible for chlorine activation and ozone depletion taking place on Polar Stratospheric Clouds (PSCs), but ozone loss also occurs on sulfate aerosols, and this is evident following volcanic eruptions (Solomon, 1999; Stenchikov et al., 2002). Ozone depletion depends upon a complex interaction between meteorological effects (for example temperature of the polar vortex, frequency and occurrence of sudden warmings), stratospheric photochemistry and, critically, halogen concentrations connected with the release of CFCs in the last few decades. Reductions in ozone column following Pinatubo of 2% in the tropics and 5% in higher latitudes were observed when particle Surface Area Densities (SAD) exceeded $>$10 $(\mu\text{m})^2/\text{cm}^3$ (e.g. Solomon, 1999). Rasch et al. (2008) noted regions with high aerosol SAD associated with geoengineering sulfate aerosol were coincident with cold temperatures (see figure 4) and indicated concern that ozone depletion might be possible, at least until most active chlorine has been flushed from the stratosphere (thought to occur after about 2050). Recently, Tilmes and colleagues have begun to explore some aspects of ozone depletion associated with geoengineering, and we summarize some of that work here.

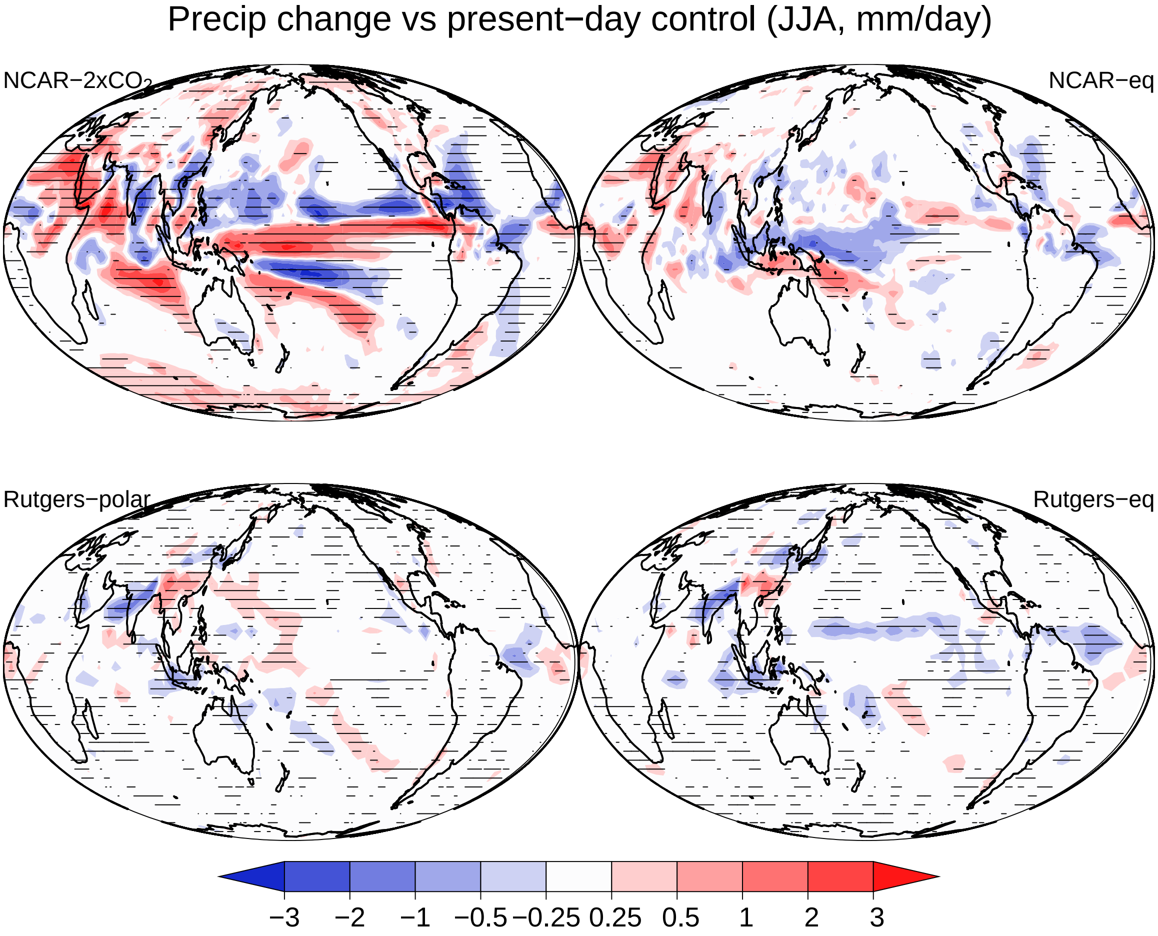

Figure 6: Change in precipitation associated with perturbations to greenhouse gases and geoengineering for two models during the June, July, August months: Top row shows differences between present day and doubling of CO_2 in the NCAR model CCSM using a slab ocean model. The top left panel shows the changes induced by $2xCO_2$. Top right panel shows the additional effect of geoengineering (with a 2 Tg S/yr source). Bottom row shows the precipitation changes for the GISS model using an A1B transient forcing scenario and full ocean model (between 2020-2030) with geo-engineering. Left panel shows the changed distrubution using 1.5 Tg S/yr injection at 68N. Bottom right panel shows the change introduced by a 2.5 Tg S/yr injection in the tropics. Hatching shows areas where difference exceed two standard deviations of an ensemble of samples from a control simulation.

Tilmes et al. (2007) estimated Arctic ozone depletion for the 1991-92 winter following the eruption of Mt Pinatubo based on satellite observations, aircraft and balloon data, and found enhanced ozone loss in connection with enhanced SAD. They used an empirical relationship connecting meteorological conditions and ozone depletion to estimate 20-70 DU extra ozone depletion from the volcanic aerosols in the Arctic for the two winters following the eruption.

Tilmes et al. (2008b) estimated the impact of geo-engineered aerosols for future halogen conditions using a similar empirical relationship, but this time including aerosol loading and changing halogen content in the stratosphere. They based their estimates of ozone depletion on an extrapolation of present meteorological condition into the future, and assumptions about the amount and location of the geoengineering aerosol. They predicted a substantial increase of chemical ozone depletion in the Arctic polar regions, especially for very cold winters, and a delay of 30-70 years of the recovery of the Antarctic ozone hole.

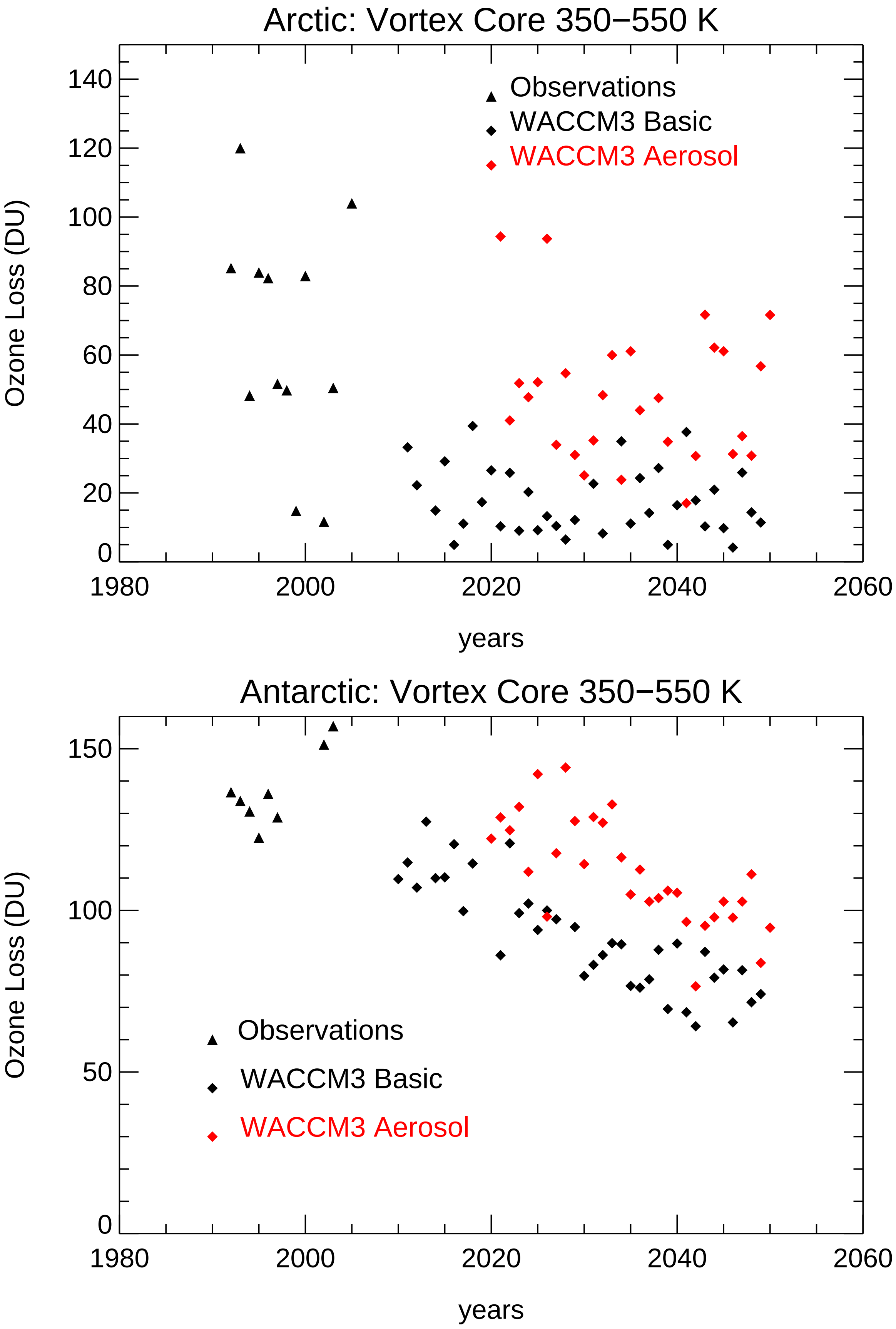

Figure 7: Partial chemical ozone depletion between 350 and 550 K in the Arctic vortex core up to April (top panel) and in the Antarctic vortex core by mid October (bottom panel), derived using the baseline model run (black diamonds), the volcanic aerosol model run (red diamonds) and observations (Tilmes et al, 2006), black triangles.

Tilmes et al. (2008a) extended their previous calculation by using one of the aerosol distribution calculated in Rasch et al. (2008) to explore the impact of geo-engineered sulfate aerosols. Rather than estimating ozone depletion using the empirical relationships the study used the interactive chemistry climate model WACCM (Whole Atmosphere Chemistry Climate Model). The configuration included an explicit representation of the photochemistry relevant to the middle atmosphere, (Kinnison et al., 2007) and SOM to allow a first order response of the troposphere to greenhouse warming, and assess changes to the middle atmosphere chemical composition and circulation structures, and the interaction between the chemistry and dynamics.

Two simulations of the time period 2010 to 2050 were performed; 1) a baseline run without geoengineering aerosols, and 2) a simulation containing geoengineering aerosols. For the baseline run, monthly mean background values of aerosols were assumed to match background SAGEII estimates (SPARC, 2006). For the geoengineering run, a repeating annual cycle of aerosols derived from the *volc2* scenario of Rasch et al. (2008) was employed. That scenario assumed aerosols with a particle size distribution similar to that following a volcanic eruption, and aerosol burden produced from a 2 Tg S/year injection of SO_2. Both model simulations used the IPCC A1B greenhouse gas scenario and changing halogen conditions for the stratosphere. In the model simulations the halogen content in the stratosphere was assumed to decrease to 1980 values by around 2060 (Newman et al., 2006). The study thus explored the impact of geo-engineering during a period with significant amount of halogens in the stratosphere so that ozone depletion through surface chemistry is important.

Beside the desired cooling of the surface, and tropospheric temperatures, enhanced sulfate aerosols in the stratosphere directly influence middle atmosphere temperatures, chemistry and wind fields. The increases of heterogeneous reaction rates in the stratosphere affect the amount of ozone. Ozone plays an important role in the energy budget of the stratosphere, absorbing incoming solar energy, and outgoing energy in the infrared. It therefore influences temperatures (and indirectly the wind field), especially in polar regions. Additional aerosol heating also results in warmer temperatures in the tropical lower stratosphere (between 18 and 30 km). This results in an increase of the temperature gradient between tropics and polar regions (as mentioned in Robock (2000)). As a consequence, the polar vortex becomes stronger, colder, and the Arctic polar vortex exists longer with geoengineering than without, which influences polar ozone depletion.

In the tropics and mid-latitudes enhanced heterogeneous reactions cause a slight increase of ozone due to the shift of the NO_x/NO_y equilibrium towards NO_y in the geoengineering run (around 2-3% maximum around 20-30 degrees North and South). In polar regions an increase of heterogeneous reaction rates have a more severe impact on the ozone layer.

Chemical ozone loss in the polar vortex betweeen early winter and spring can be derived for both model simulations. These results can be compared to estimates derived from observations between 1991-92 and 2004-05 for both hemisphere Tilmes et al. (2006, 2007). Such results are displayed in Figure 7. Estimates for present day depletion is indicated in black triangles. Estimates for the control simulations, and geoengineered atmosphere are shown in back and red diamonds respectively.

The WACCM model does a relatively good job of reproducing the ozone depletion for the Antarctic vortex (bottom panel). Ozone loss decreases linearly with time (black diamonds), and year to year variability in the model is similar to that of the observations. The WACCM model suggests a 40-50 DU increase in ozone depletion in the Antarctic Vortex due to geoengineering.

The model reproduces the depletion and variability much less realistically in the Arctic (top panel). Averaged temperatures in the simulated vortex are similar to observations, but the model does not reproduce the observed chemical response. The simulated polar vortex is $\sim$2-5 degrees too small and the vortex boundary is not as sharp that seen in the observations. The ozone depletion starts later in the winter due to warmer temperatures in the beginning of the winter and there is less illumination at the edge of the smaller vortex (necessary to produce the depletion). Chemical ozone depletion for the WACCM3 baseline run in the Arctic is less than half that derived from observations. Underestimates of Bromine concentrations may also cause the underestimation of chemical ozone loss.

Examples of spatial changes in ozone depletion are shown in Figure 8, which displays the difference between

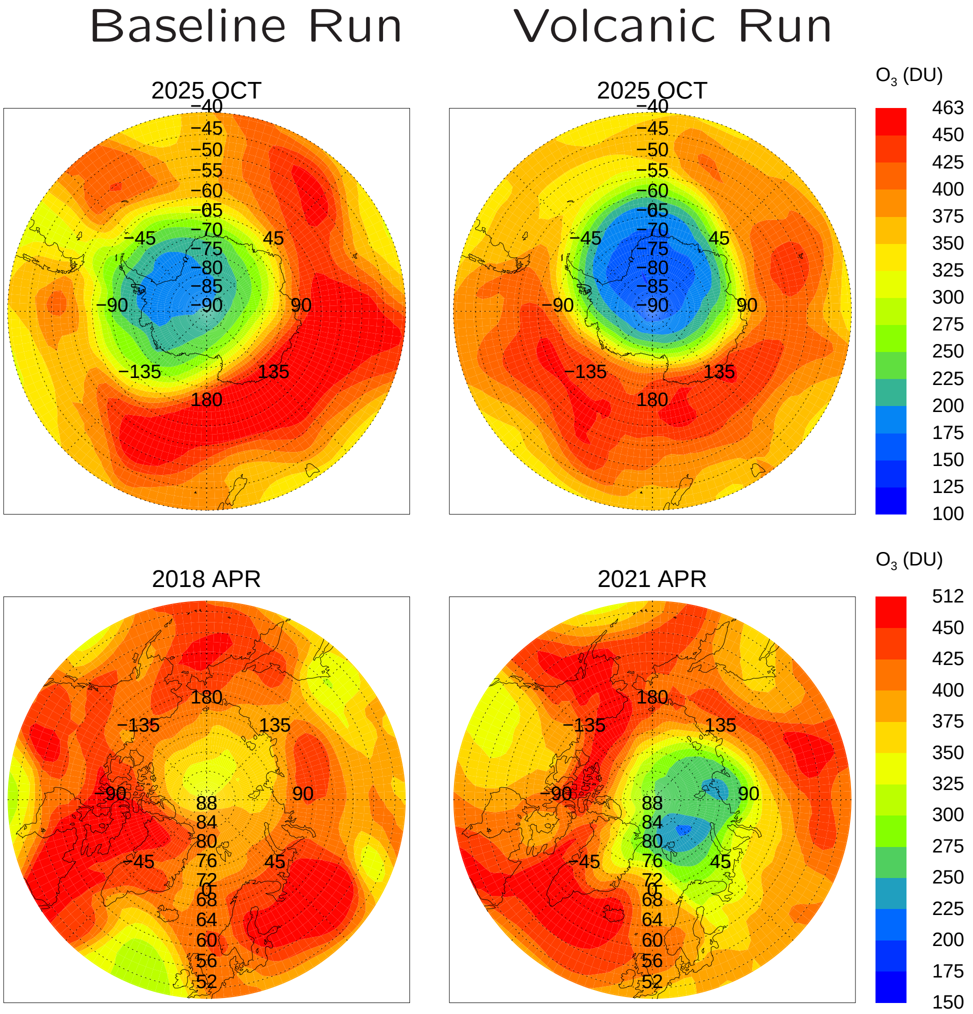

Figure 8: Column ozone for baseline run (left column) and geoengineering run (right column) for two meteorologically similar Antarctic winters in mid-October 2025 (top panel) and the coldest simulated Arctic winters in the beginning of April. (DU= Dobson units)

the baseline and geoengineering runs for Antarctica (top panel) for two winters with similar temperature structure and for the coldest winter of each simulation in the Arctic (bottom panel). Antarctic winters show ∼30 DU smaller column ozone values for the geoengineering simulation. Larger ozone losses occur over a wider area of the vortex for the geo-engineering model run.

The geoengineering simulation suggests that ozone depletion will be somewhat larger in the Arctic. The amplitude of the variability in ozone depletion is increased in the geoengineering simulation, and colder vortex temperatures occur during winter and spring. The coldest three winters of the geo-engineering run are 1 - 2.5 degrees colder than the coldest winter in the baseline run (between 20-25 km) between mid-December and March. The warm Arctic winter in the baseline case show little ozone depletion, (Figure 8, bottom left). The colder temperatures and larger vortex in the geoengineering run result in increased depletion compared to the control. The Arctic ozone column falls below 250 DU in the vortex core and reach latitudes of 70 degrees North. *N.B.* The inability of the model to reproduce the chemical signal of observed ozone depletion in the unperturbed calculation means one must be cautious in interpreting the model estimates for the Arctic.

The Tilmes et al. (2008a) study found that the Antarctic chemical ozone loss would be 40-50 DU larger with geoengineering than without, and that there would be a a delay of 20-30 years before the ozone recovery produced by the gradual depletion of halogens would begin again.

The Tilmes et al. (2008b) and Tilmes et al. (2008a) studies therefore indicate that polar ozone depletion from geoengineering could occur during this century and that geoengineering may have a significant impact on the ozone layer.

4 Summary, Discussion, Conclusions

We have reviewed geoengineering by stratospheric aerosols as a possible means of mitigating the climate change associated with increased greenhouse gases. Sulfate aerosols in the stratosphere will increase the refectivity of the planet and counteract some of the effects of CO_2 warming. Part of the attraction of using stratospheric aerosols arises because volcanic eruptions form a natural but imperfect analog to geoengineering. History has demonstrated that sulfate aerosol, in sufficient amounts, will cool the planet, and that the earth system can survive this kind of perturbation. Although the topic has been discussed over the last 30 years, only very recently have attempts been made to understand the interactions between various components of the climate system using modern tools for understanding climate consequences. These tools provide opportunities to quantify the interactions and consequences, and to explore those consequences on time scales that are much longer than the influence of a single volcanic eruption.

We have shown that sophisticated models used to simulate the Earth system produce the intended physical response, that is, the earth does cool, and many components of the system return to a state more like an unperturbed earth.

While it is tempting to become enthusiastic about the method. We hope that this study also tempers that enthusiam to some degree for the following reasons:

- We have shown that the delivery of aerosols or their precursors is a formidable task. It would take order a million flights of four hour duration (2500 km) per year to deliver the nominal amount of aerosol (10 Tg particles /yr = 2.5 Tg S /yr) needed to balance the warming associated with increasing greenhouse gas emissions. These numbers are still quite rough, and it is possible that an increase by a factor of 2-4 might be required. The issues associate with delivery are relevant to other proposed delivery systems (artillery, balloons, etc.) although details will certainly be different.

- Although it is possible to cool the earth to approximately the same globally averaged surface temperature, it is not likely that all aspects of the physical system will return to a state like that prior to human induced CO_2 increases. It is important to emphasize the uncertainties in our characterization of these issues. We have made initial exploratory forays into understanding the consequences of geoengineering, but much work remains to be done. The high sensitivity of polar regions to processes regulating energy in and out of the system would make it difficult to reproduce precisely the seasonal cycle of the polar climate for a preindustrial (or even present day) world with geoengineering.

  A recent study by Stenchikov et al. (2006) showed that models have difficulty capturing the regional response of the climate system to volcanic eruptions. They argued that volcanoes' influence on the Artic Annular Oscillation is associated with the extra heating in the equatorial lower stratosphere, changing the temperature gradient in the lower stratosphere vortex, producing stronger westerlies and a winter warming over northern Eurasia and N. America. Models identified in that paper (that participated in the IPCC) tended to underestimate, and misplace the northern hemisphere winter surface temperature warming seen over Siberia in observations following an eruption. This suggests that while the zeroeth order response of a surface cooling is likely to be robust, the first order response of other components of the climate systems is a difficult problem and that model regional responses to stratospheric forcing changes must be viewed cautiously.

As discussed in section 3.4, there are also hints that rainfall patterns would be different from an undisturbed earth, although it is likely that they would be much closer to that desired realm than in a world with 2xCO_2 and no geoengineering.

- We have shown that an increase in aerosol burden is likely to increase ozone depletion. We have shown that current chemistry climate models have difficulty in reproducing quantitatively the dynamics and chemistry of the arctic middle atmosphere. Better coupled chemistry climate models would allow a better estimate of ozone, sulfate aerosol, dynamical interactions. The first step is to improve the models capability in reproducing present day ozone representation, particularly for the northern hemisphere.

- Reductions in ozone will lead to increases in solar ultraviolet-B radiation reaching the Earth's surface with a potential impact on human health (Madronich and de Gruijl, 1993; Ambach and Blumthaler, 2005), and biological populations (Blaustein et al., 1994).

  The increase in UV associated with ozone depletion could be compensated for by increased extinction and attenuation by the aerosol cloud itself. Vogelmann et al. (1992) and Wetzel et al. (2003) explored the compensation between these effects. Vogelmann et al. (1992) studied the effect for volcanic eruptions and concluded that for stratospheric aerosol optical depths of 0.1-0.2 (approximately the value required for geoengineering), that ozone and aerosol effects approximately compensated. At higher aerosol amounts the aerosol attenuation did not balance the ozone enhancement, and UV was enhanced at the surface. Those calculations should be repeated with a focus upon geoengineering and global warming, since ozone distributions and aerosol spatial and particle size distributions might differ significantly for geoengineering scenarios compared to their volcanic eruption calculations.

- Gu et al. (2003) showed that volcanic aerosols from the Pinatubo eruption substantially increase diffuse radiation worldwide, with a resulting enhancement to photosynthesis and uptake of CO_2. The same effect is to be anticipated with the geoengineering shield. Govindasamy et al. (2002) explored some aspects of interactions between the physical earth system and the biosphere. They showed that stabilizing the temperature but not CO_2 induced a change in Net Primary Productivity. Their study had a number of limitations: 1) they used a prescribed CO_2 concentration, eliminating important feedbacks; 2) they did not use a biospheric model that included nutrient limitation; 3) They did not include an ocean biosphere; 4) their model had no species competition; 5) their model was not sensitive to changes in the ratio of direct to diffuse radiation.

  While we know that ecosystems survive occasional volcanic eruptions, it is not clear what the consequences would be to long term changes in direct/diffuse energy, or in increases in UV radiation. These issues argue for more attention on the consequences of stratospheric aerosols to ecosystems.

  We mention in passing that the change in ratio of direct to diffuse radiation will also have an effect on solar energy production with technologies that make use of solar concentrators. Advances in solar energy production that operate efficiently in the presence of diffuse radiation are also possible, but a different technology is needed. Characterizing the consequences of geoengineering to these technologies is worthwhile.

- As mentioned in section 2, 3.2 and 3.4, larger aerosol particles do absorb in the longwave. The scattering of incoming solar energy is thus partly compensated for by the absorbtion in the longwave. The proclivity of this geoengineering method to form large particles makes it a less efficient solution than it would be if small particles were easily generated.

- There are also occasional concerns voiced about increases to acid rain from this type of geoengineering. We have shown that although the perturbations to the stratospheric sulfur cycle are quite large (increasing the background sources there by a factor of 15-30), they are perhaps 2% of the total (troposphere+stratosphere) sulfur sources. We believe therefore that an impact on acid deposition from geoengineering is unlikely, and could easily be balanced by reductions in emissions important to the troposphere, unless the deposition occured on a region that normally sees little sulfate deposition (perhaps the poles).

- It is obvious that our models of the sulfur cycle could be substantially improved. It would be desirable to move beyond the “bulk” aerosol formulations used here to models that included the evolution of the particle size distribution, accounting explicitly for aerosol growth and coagulation. This would include a mechanism to move from the source as determined by the delivery system, to evolution within the plume and finally to scales resolved by a global model.

- It is clear that this geoengineering method will not alleviate the problems engendered by absorption of CO_2 in the oceans, with a resulting decrease on ocean pH.

We close by stating that substantial reductions in greenhouse gas emissions must take place soon to avoid large and undesirable climate impacts. The first response of society to this evidence ought to be to reduce greenhouse gas emissions. A second step might be to explore what is deployable in a planetary emergency to mitigate some of the effects of a planetary warming. We emphasize that while the studies highlighted here are a step along the way, we believe no proposal (including the ideas explored here) has yet completed the series of steps required for a comprehensive and thoroughly studied geoengineering mitigation strategy occuring in the peer reviewed literature (Cicerone, 2006). Our studies of geoengineering by sulfate aerosols suggest it will ameliorate some consequences of global warming. The study highlights some positive aspects of the strategy. However many uncertainties remain in understanding the influence of geoengineering on the climate system (particularly on aspects related to likely impacts on the biosphere). More work is required to understand the costs, benefits, and risks involved, and to reconcile the legal, political and ethical issues of geoengineering.

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