



Exploring the geoengineering of climate using stratospheric sulfate aerosols: The role of particle size

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[1] Aerosols produced in the lower stratosphere can brighten the planet and counteract some of the effects of global warming. We explore scenarios in which the amount of precursors and the size of the aerosol are varied to assess their interactions with the climate system. Stratosphere-troposphere exchange processes change in response to greenhouse gas forcing and respond to geoengineering by aerosols. Nonlinear feedbacks influence the amount of aerosol required to counteract the warming. More aerosol precursor must be injected than would be needed if stratosphere troposphere exchange processes did not change in response to greenhouse gases or aerosols. Aerosol particle size has an important role in modulating the energy budget. A prediction of aerosol size requires a much more complex representation and assumptions about the delivery mechanism beyond the scope of this study, so we explore the response when particle size is prescribed. More aerosol is required to counteract greenhouse warming if aerosol particles are as large as those seen during volcanic eruptions (compared to the smaller aerosols found in quiescent conditions) because the larger particles are less effective at scattering incoming energy, and trap some outgoing energy. About 1.5 Tg S/yr are found to balance a doubling of CO₂ if the particles are small, while perhaps double that may be needed if the particles reach the size seen following eruptions. **Citation:** Rasch, P. J., P. J. Crutzen, and D. B. Coleman (2008), Exploring the geoengineering of climate using stratospheric sulfate aerosols: The role of particle size, *Geophys. Res. Lett.*, 35, L02809, doi:10.1029/2007GL032179.

1. Introduction

[2] A resurgence of interest in geoengineering (the deliberate change of the Earth's climate by mankind [e.g., Keith, 2000]) is occurring from a concern 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. The assessment by the Intergovernmental Panel on Climate Change [Intergovernmental Panel on Climate Change (IPCC), 2007] 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 a second step might be to explore strategies to mitigate some of the planetary warming. Two recent papers [Crutzen, 2006; Wigley, 2006] explored a geoengineering idea going back to Budyko [1974], who speculated that a deliberate production of stratospheric aerosols might increase the planetary albedo, and cool the planet, ameliorating some (but not all) of the effects of increasing CO₂ concentrations.

[3] 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.

[4] This study explores the consequences of variations in the amount and size of the particles generated by injecting precursors of sulfate aerosols into the stratosphere on the climate system. 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; Crutzen, 2006] address some of these issues and remind us that this approach does not treat all the consequences of higher CO₂ concentrations (such as ocean acidification).

2. Model

[5] We use a coupled Atmosphere Ocean General Circulation Model (AOGCM) variant of the NCAR Community Atmosphere Model (CAM3) [Collins *et al.*, 2006], configured to use a finite volume (FV) solution for atmospheric dynamics [Lin, 2004; Rasch *et al.*, 2006] coupled to a slab ocean model (SOM) [Kiehl *et al.*, 2006]. The model has 52 layers from the surface to 80 km, with a horizontal resolution of 1.9 degrees latitude by 2.5 degrees longitude, producing a reasonable climate for the troposphere and middle atmosphere [Sassi *et al.*, 2002]. SOM heat fluxes were calculated using the strategy of Kiehl *et al.* [2006]. The use of a SOM with a thermodynamic sea ice model precludes a dynamic response from the ocean and sea-ice, which would require a more complex model.

[6] We explore the climate system forcing and response associated with "direct" aerosol forcing. CAM3 normally uses five classes of aerosol (sulfate, soot, organic carbon-

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Table 1. Emissions, Burdens, Residence Times, and the Equilibrium Temperature Change^a

Case	CO ₂ Forcing, ppmv	Geosulfate Source, Tg S/yr	Size	Burden, Tg S	Residence Time, years	ΔT_s , K
CONTROL	355	none	—	—	—	0
2xco2	710	none	—	—	—	2.1
bg1	355	1 Tg S/yr	small	2.7	2.7	-1.7
volc2	355	2 Tg S/yr	large	5.9	3.0	-2.0
bg1co2	710	1 Tg S/yr	small	2.4	2.4	0.9
bg2co2	710	2 Tg S/yr	small	5.5	2.7	-0.8
volc2co2	710	2 Tg S/yr	large	5.1	2.5	0.7

^aThe “large” volcanic-like particle distribution, is labelled “volc.” Cases using “small” background-like particle distribution, are labelled “bg.” See text for description of small and large particle definitions.

ceous aerosol, sea salt and dust). The model can use a prescribed, repeating annual cycle for the aerosol, or can predict them from production, transport and loss processes. These simulations use prescribed aerosols, except in some runs we introduce an additional class of aerosol we call ‘geosulfate’ using the predictive mode with an explicit formulation of stratospheric sulfate production, transport and loss. The geosulfate is assumed to arise from injection of H₂S or SO₂ into the lower stratosphere. It evolves interactively with the model climate. The model responds to the direct radiative forcing from both the prescribed and predicted aerosols. We assume non-interacting external mixtures, and neglect aerosol/cloud interactions and reactive photochemistry.

[7] Our geosulfate parameterization follows *Barth et al.* [2000], with additional processes appropriate to the stratosphere [*Stratospheric Processes and Their Role in Climate (SPARC)*, 2006]. Sulfate is produced through oxidation of SO₂ and lost via scavenging in the troposphere. The geosulfate is assumed to be 75% H₂SO₄ and 25% water by weight. SO₂ is oxidized to gaseous H₂SO₄ which condenses to form drops that are transported and sediment. The aerosol evaporates when it becomes subsaturated, and is photolyzed to SO₂ where it can again oxidize. We use a “bulk” formulation carrying only the aerosol mass (the particle size distribution is prescribed). Modeling the evolution of the size distribution would require a more complex framework and the outcome depends upon many factors, including (among other things) details of the delivery mechanism for precursors, the relative rates of dilution and transformation and the partial pressure of H₂SO₄ gas. These processes occur on sub-grid scales and require a complex formulation. Since we do not model the particle growth we have explored scenarios spanning much of the size range that the aerosols might attain, assuming the distribution will either be “small”, like that seen during background situations with volcanically quiescent conditions [*Bauman et al.*, 2003], or “large” like that seen 6–12 months after a volcanic eruption [*Stenchikov et al.*, 1998]. Our small particles are assumed to follow a log-normal distribution with a dry mode radius, standard deviation, and effective radius of 0.05/2.03/0.17 μm respectively. Corresponding numbers for the large volcanic-like distribution are 0.376/1.25/0.43 μm respectively [*Collins et al.*, 2004]. When the particles are small they primarily scatter in the solar part of the energy spectrum, and play no role in the longwave; when large they scatter and absorb in the solar wavelengths, but also absorb in the infrared [*Stenchikov et al.*, 1998]. The

size of the aerosol thus has a strong influence on the climate.

3. Simulations

[8] *Crutzen* [2006] used the surface temperature (T_S) response to Pinatubo to estimate that 1–2 Tg S/yr would counteract the T_S response to a doubling of CO₂. *Wigley* [2006] used an energy balance model to conclude that ~ 5 Tg S/yr in combination with emission mitigation would suffice. *Govindasamy and Caldeira* [2000] and *Matthews and Caldeira* [2007] mimicked the impact of stratospheric aerosols by reducing the solar constant to diminish the energy entering the atmosphere (by 1.8%). We have extended these studies to include geoengineered aerosol evolution, feedback, and response.

[9] We evaluate six simulations against a control. In the geoengineering cases SO₂ was injected uniformly and continuously in a 2 km thick region at 25 km between 10°N and 10°S. The non-control experiments were run for 50 years, and the last 20 years averaged. The control was run for 140 years. The first 20 years were ignored, and the balance divided into six 20-year samples, which were used to produce an ensemble average and standard deviation (SD). We focus only upon features where differences exceed 2 SD. Table 1 shows emissions, burdens, residence times (defined as the burden/source at equilibrium) and the equilibrium temperature change.

[10] This version of CCSM produces a T_S increase of 2.1 K for a doubling of CO₂, on the low end of the spectrum of climate models [*IPCC*, 2007]. The low sensitivity is due to our choice of the FV dynamics module mentioned above. *Matthews and Caldeira* [2007] showed that the geoengineering forcing required to neutralize a CO₂ increase is essentially independent of the climate sensitivity.

[11] Case *volc2* (2 Tg S/yr of large particles at present day CO₂) produces a 2.0 K cooling. If the system were linear the combination of the two forcings would result in a T_S 0.1 K warmer than present day. The combination of the two forcings (case *volc2co2*) leaves the planet 0.7 K warmer than present day, a discrepancy of 0.6 K compared to the sum of the individual responses. The nonlinear response indicates feedbacks resulting from enhanced stratosphere-troposphere exchange (STE) [*Butchart and Sciafe*, 2001; *Rind et al.*, 2001], reducing the residence time in a doubled CO₂ climate. A similar result is seen when 1 Tg S/yr is injected assuming “small” aerosol particles. If the injection takes place with present day greenhouse gases (case *bg1*) a 1.7 K cooling is seen, but in a doubled CO₂ world (case

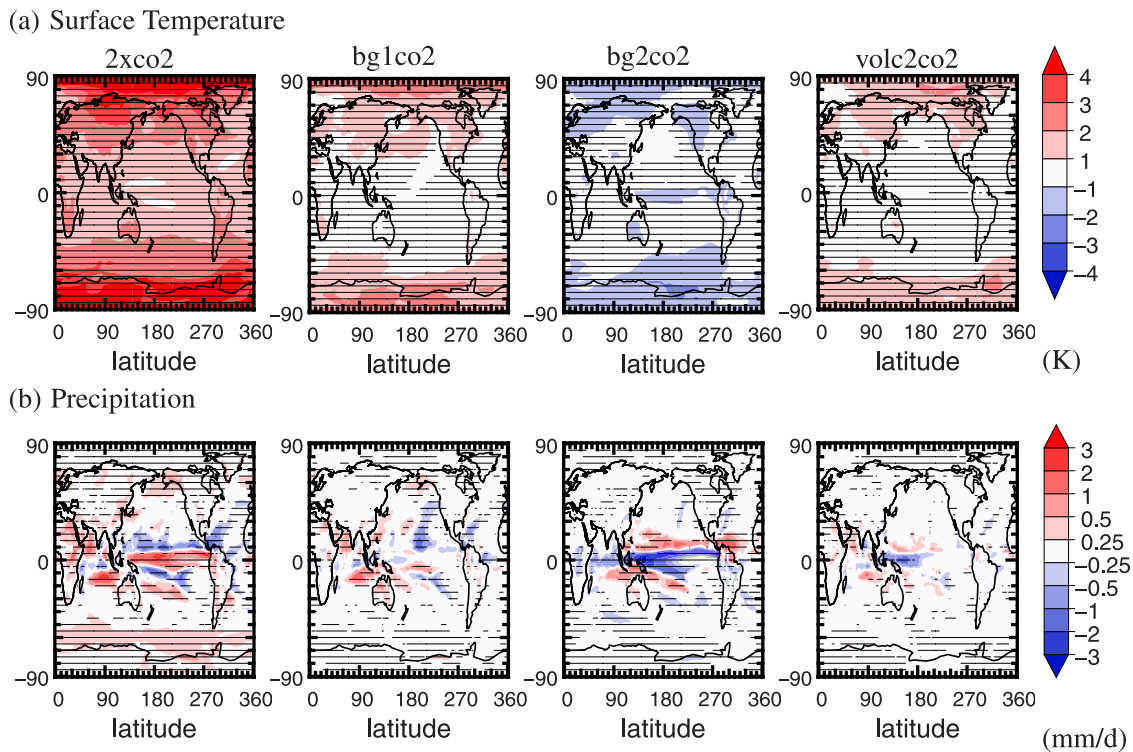


Figure 1. Change in (top) annual averaged T_S and (bottom) precipitation between experiments and the ensemble average of the present day (control) simulation. Hatched regions show areas where differences exceed two standard deviations of the ensemble.

bg1co2) the combined forcing results in a globally averaged T_S 0.9 K warmer than present day, rather than 0.4 K warmer, a discrepancy of 0.5 K. To produce a simulation in which the geosulfate is sufficient to counterbalance the greenhouse warming, we have also explored a 2 Tg/yr emission scenario assuming small particles (case *bg2co2*). This overcools the planet to ~ 0.8 K below present day. A source of ~ 1.5 Tg S/yr (assuming small particles and that the discrepancy associated with the dynamical response is consistent) would produce a T_S within 0.1 K of present day.

[12] Changes in annual averaged T_S and precipitation for some scenarios are shown in Figure 1. Hatching indicates where changes exceed 2 SD (likely to be statistically important). The *2xco2* case shows the familiar model response to a doubling of CO_2 with enhanced warming in polar regions, particularly over land [IPCC, 2007]. Injection of 1 Tg S/yr as small particles (*bg1co2*) reduces the warming equatorward of 40 degrees to <1 K, and reduces the enhanced precipitation from 3 mm/day in the tropics to ~ 1 mm/day. Precipitation changes over continental regions are small except over Australia. T_S differences still exceed 2 K near the poles. Increasing geosulfate production to 2 Tg S/yr as small particles (*bg2co2*) produces a global cooling with a larger response over continents than oceans. The rightmost panel shows the climate response to a 2 Tg/yr source of larger particles. Largest perturbations from present day again occur over continents. The reduced single scattering albedo of the larger particles and increased absorption in the infrared lessen the impact of the geoengineering, making large particle geosulfate less effective in cooling the planet.

[13] Figure 2 shows changes in T_S and sea ice extent for cases *bg1co2* and *bg2co2* with season. The seasonal cycle amplitude is strongly modulated by geosulfate. Winters are a few K warmer than present day when the production rate is too small to counteract the greenhouse warming (displayed for case *bg1co2* and not shown for *volc2co2*). Case *bg2co2* has cooler winters than present day. A precise choice of geosulfate might produce a seasonal cycle like today's, but it is clear that the polar region is sensitive to subtleties of the geoengineering. Sea ice extent decreases for *2xco2* by as much as 20% in the arctic during summer (not shown). Cases *volc2co2* (not shown) and *bg1co2* counteract this reduction, with a small area in the NH where ice fraction is still 10% less than present day. Larger aerosol burdens (case *bg2co2*) overdo the globally averaged cooling, producing more ice in the southern hemisphere higher than present day. An analysis of runs using a version of CCSM discussed by Kiehl *et al.* [2006] suggests that changes in sea ice due to doubling of CO_2 are underestimated in a SOM, when compared to transient coupled simulations that include a dynamical sea ice and ocean model that has a relatively realistic simulation of the seasonal evolution of sea ice [Holland *et al.*, 2006]. These factors suggest our analysis will only provide hints to the changes in sea ice.

[14] Zonal averages for temperature change, geosulfate mixing ratio and Surface Area Density (SAD) are shown in Figure 3. Case *2xco2* shows a 2 K warmer troposphere and (as much as 10 K) cooler stratosphere. Geoengineering cools the troposphere, but does not ameliorate the stratospheric cooling. The tropical region from 10–20 km shows

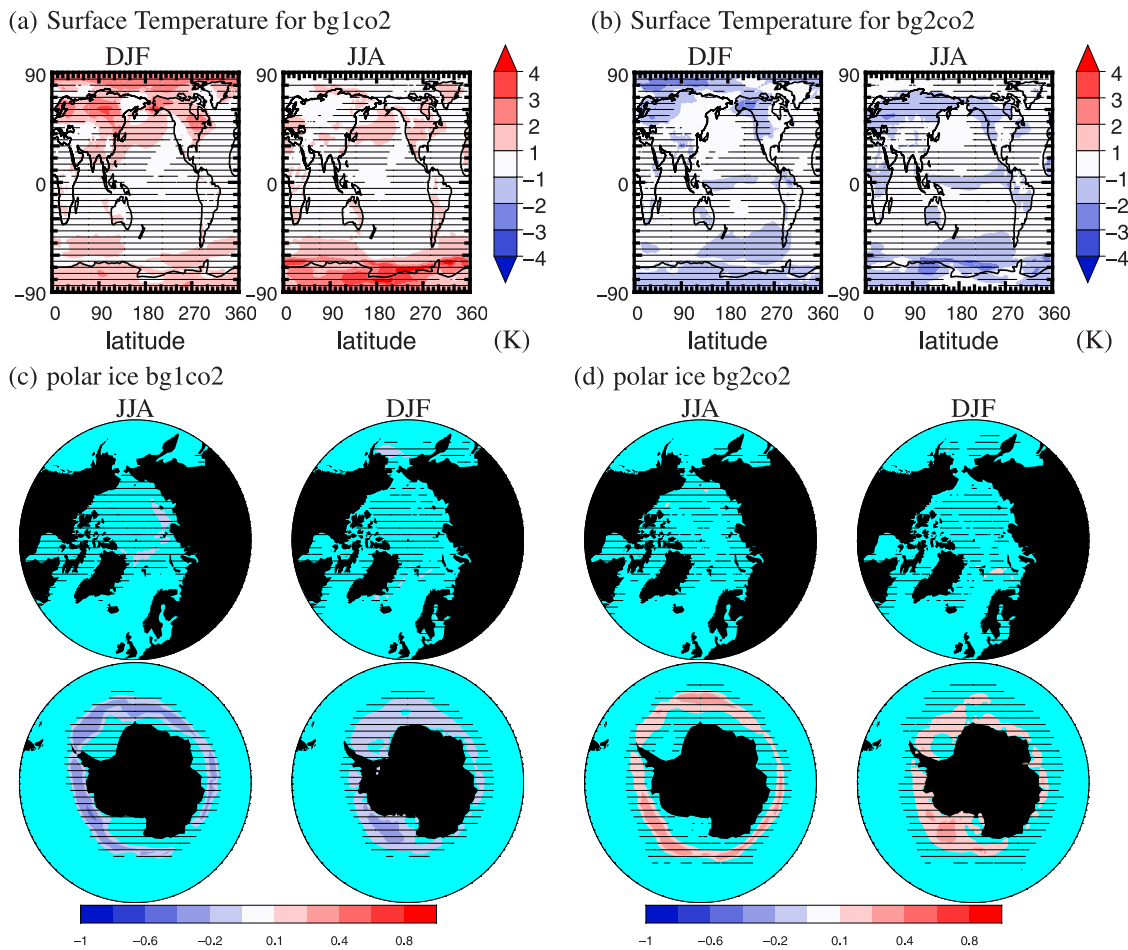


Figure 2. Changes to surface temperature and sea ice as a function of season. Hatching as in Figure 1.

a delicate balance most sensitive to the geosulfate forcing compared to other parts of the atmosphere, which return to present-day conditions more readily. Highest mixing ratios are found in the tropics near the altitude of injection. Mid and high latitudes show a strong seasonal variation in amount and altitude, driven by the residual mean circulation, and more vigorous exchange between stratosphere and troposphere in the winter hemisphere. Above 35 km the sulfate aerosol evaporates and photolyzes back to SO_2 [SPARC, 2006]. Mixing ratios are larger in the summer hemisphere. SADs are highest at the winter pole, where the air densities are higher at the altitude of the aerosol maximum. SAD is important for stratospheric ozone photochemistry, and is the quantity typically produced by aerosol retrievals [e.g., Bauman *et al.*, 2003]. SADs using the “small particle assumption” are as high as those seen during a moderate eruption like Pinatubo. Since the surface area is proportional to aerosol mass over r_{eff} , and the mass of geosulfate is relatively insensitive to particle size, the SAD for a large particle injection (case *volc2co2*) are a factor of three lower than seen in Figure 3.

[15] The surface chemistry responsible for chlorine activation and ozone depletion has focused most attention on Polar Stratospheric Clouds (PSCs), but ozone loss also occurs following volcanic eruptions [Robock, 2000]. Ozone depletion on sulfate aerosols is subject to saturation effects

[Fahey *et al.*, 1993] and it is unclear what influence geosulfate may play. The red contour lines on Figure 3 indicate regions cold enough that ozone depletion may be important. Reductions in ozone column following Pinatubo of 2% in the tropics and 5% in higher latitudes were observed when $\text{SAD} > 10 (\mu\text{m})^2/\text{cm}^3$ [Robock, 2000]. These areas coincide, so ozone depletion from geosulfate is possible until most active chlorine has been flushed from the stratosphere (after 2050). Because PSCs also occur in these cold regions there may be little additional impact of the geosulfate aerosol.

4. Summary

[16] We have examined the role that sulfate aerosols in the stratosphere have on brightening the planet and counteracting some of the effects of CO_2 warming. Six scenarios were used to explore the climate system response to geosulfate amount and size.

[17] While the radiative forcing from geosulfate is higher in equatorial regions, the largest response occurs at the poles, consistent with the general behavior of climate models to uniform forcing [IPCC, 2007] and the response to volcanic eruptions [Robock, 2000]. Stratosphere Troposphere Exchange (STE) processes respond to greenhouse gas forcing and interact with geosulfate. Nonlinear

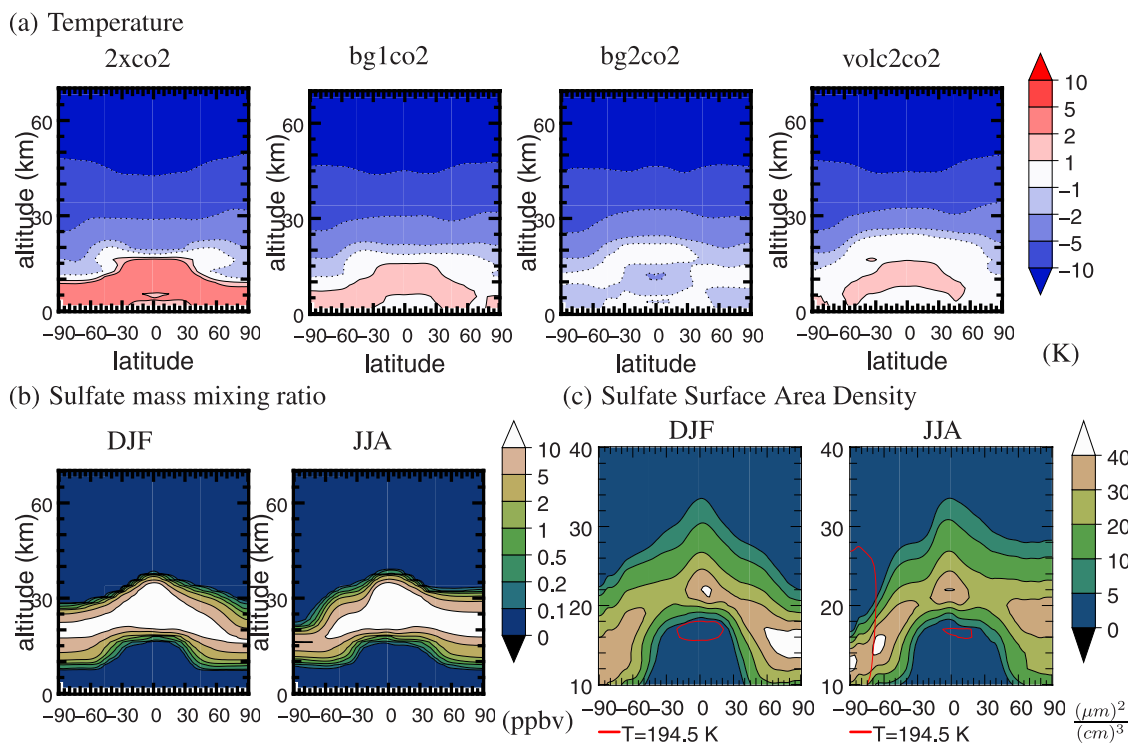


Figure 3. (top) Zonal average of annually averaged temperature. (bottom) The seasonal aerosol mixing ratio and surface area for June, July, August (JJA) and December January February (DJF) for experiment *bg2co2* (see Table 1). The red contour shows the 194.5 K temperature isotherm.

feedbacks modulate these processes and influence the amount of aerosol precursor required to counteract CO_2 warming. We find 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 also plays a role. While the physics of aerosol formation and growth is understood, particle size is governed by a complex array of processes and influenced by the strategy employed to deliver the aerosol precursors to the stratosphere, making it difficult to predict the aerosol size distribution. Therefore we have explored the impact of a range of prescribed particle sizes. 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 is more than enough to balance the warming in global-mean terms from a doubling of CO_2 if particles are small, but insufficient if the particles are large. Small particles are optimal for geoengineering through radiative effects, but also provide more surface area for chemistry to occur. As also given by Govindasamy and Caldeira [2000], the seasonal cycle of the polar climate is quite sensitive to the amplitude of the forcing by the geoengineering aerosol. A partial compensation by geosulfate will result in warmer winters than present day.

[18] The coincidence of aerosol surface area at temperatures low enough for ozone depleting photochemistry indicates that chemical studies ought be considered. With present chlorine loadings the impact on ozone is likely to be small because most of the ozone in cold regions is already

depleted. As chlorine levels decrease, the role of the surface chemistry will become less important and the presence of the geosulfates may be less important, but quantitative statements require a comprehensive photochemical treatment.

[19] We close with a reminder that our study has examined only the physical response of the climate system to geoengineering. Other components (e.g. ecosystems, ocean circulations, and the cryosphere) requiring more complex models than used here should be considered.

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