Title: Effects of stratospheric sulfate aerosol geo-engineering on cirrus clouds

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Abstract:

Cooling the Earth through the injection of sulphate into the stratosphere is one of the most discussed geo-engineering (GE) schemes. Stratospheric aerosols can sediment into the troposphere, modify the aerosol composition and thus might impact cirrus clouds. We use a global climate model with a physically based parametrization for cirrus clouds in order to investigate possible microphysical and dynamical effects. We find that enhanced stratospheric aerosol loadings as proposed by several GE approaches will likely lead to a reduced ice crystal nucleation rate and thus optically thinner cirrus clouds. These optically thinner cirrus clouds exert a strong negative cloud forcing in the long-wave which contributes by 60% to the overall net GE forcing. This shows that indirect effects of stratospheric aerosols on cirrus clouds may be important and need to be considered in order to estimate the maximum cooling derived from stratospheric GE.

1. Introduction

Mankind is facing a changing climate due to the emission of greenhouse gases [*Solomon et al.*, 2007]. Along with mitigation and adaptation geo-engineering (GE) has become more and more part of recent research strategies for dealing with global warming. Although the idea of influencing climate by manipulating the planetary environment is not new [*Budyko*, 1977] it has recently gained broad attention. *Shepherd et al*., [2009] gives an overview over a broad range of GE approaches and evaluates them in terms of affordability, effectiveness, timeliness and safety.

Injecting sulphate aerosols into the stratosphere belongs to the most discussed GE approaches because it is assumed to be of high effectiveness and low costs [*Shepherd et al.*, 2009; *Lenton and Vaughan* 2009, *Crutzen*, 2006; *Wigley,* 2006]. However, safety is judged to be low and more studies on the impact of stratospheric aerosols are needed in order to evaluate possible side effects. Possible side effects are impacts on the hydrological cycle, stratospheric ozone and cirrus clouds. Recent studies have addressed the question of possible effects on ozone and the hydrological cycle [*Tilmes et al.,* 2008; *Heckendorn et al.,* 2009; *Robock et al.,* 2008; *Trenberth and Dai*, 2007]. *Tilmes et al.,* [2008] analyzed the sensitivity of ozone depletion to proposed GE schemes and found that the recovery of the Antarctic ozone hole would be prolonged by some decades if large burdens of SO_2 are brought into the stratosphere on a continuous basis. Also, the hydrological cycle will likely be affected by stratospheric aerosols. *Robock et al.*, [2008] used a global climate model to investigate the regional climate responses to stratospheric sulphate injections and found modified Asian and African monsoons which reduce the precipitation regionally and thus endanger the food supply. Reduced precipitation over land was also found by *Trenberth and Dai*, [2007] who evaluated observational data after the eruption of Mt. Pinatubo in 1991.

Whereas the impacts of stratospheric sulphate injections on the ozone layer and the hydrological cycle have recently been investigated, effects on cirrus clouds remain unknown [*Robock et al.,* 2008; *McCormick et al.,* 1995, *Shepherd et al.*, 2009]. Aerosols injected into the stratosphere can enter the troposphere through sedimentation or tropopause foldings and might affect cirrus clouds through aerosol-cloud interactions [*McCormick et al.*, 1995, *Sassen*, 1992]. Several studies on the eruption of Mt. Pinatubo investigated these possible aerosol-cirrus interactions and led to a rather controversial discussion: Some studies argue that volcanic eruptions introduce larger and more abundant soluble aerosols into the upper troposphere leading to cirrus cloud formation at lower supersaturations and enhanced ice crystal number concentrations. These ideas are confirmed by ISCCP lidar measurements and detailed microphysical model simulations [*Sassen,* 1992; *Jensen and Toon,* 1992]. Other modeling studies however find only a weak aerosol effect on cirrus clouds induced by volcanic aerosols even in case of large aerosol perturbations [*Kärcher and Lohmann,* 2002; *Lohmann et al.,* 2003].

Cirrus clouds are crucial for the radiation budget of the Earth-atmosphere system. Their net radiative forcing is still difficult to determine, but it is probably positive [*Chen et al.*, 2000]. It has been shown that for mid-latitudinal cirrus clouds the combination of ice crystal number concentration, ice crystal mass and temperature of the cloud determines if the net forcing is positive or negative [*Fusina et al.*, 2007]. Thus, possible changes in the microphysical properties due to stratospheric aerosols can modify the cloud forcing. Previous studies estimating the effectiveness of stratospheric GE have not taken this side effect into account. Our motivation is driven by the concern that estimates of the effectiveness of GE lead to an over- or underestimation of the cooling because they do not consider possible indirect effects on clouds.

In this study we investigate the possible impact of stratospheric sulphate injections as proposed by several GE schemes on cirrus clouds. We perform idealized simulations in order to first study microphysical and dynamical impacts on cirrus clouds and second to examine the impact of these modified cirrus on the radiation balance and thus on the effectiveness of GE.

2. Model description

We use the general circulation model ECHAM5-HAM [*Roeckner et al.*, 2003] which is capable of simulating aerosol-cloud interactions in water, mixed-phase and cirrus clouds. It has been used to study indirect aerosol effects [*Lohmann and Ferrachat* , 2010]. In order to simulate GE in terms of stratospheric aerosols 5 Mt of gaseous SO_2 are injected per year in a band around the equator between 10°N and 10°S similar to previous studies [*Robock et al.*, 2008]. ECHAM5-HAM includes a chemistry model treating the sulfur cycle [*Feichter et al.*, 1996]. We chose to inject the SO_2 into the second lowest level of the stratosphere (corresponding to ~50-70 hPa), in order to minimize the effort and cost of this GE-strategy on the one hand and to avoid a too fast mixing with the troposphere on the other hand.

The two-moment stratiform cloud scheme for cloud droplets and ice crystals [*Lohmann et al.*, 2008] is coupled to the two-moment aerosols scheme HAM [*Stier et al.,* 2005] which predicts the aerosol mixing state as well as aerosol mass and number concentration. HAM includes the following 5 aerosol types: sulphate, black carbon, organic carbon, sea salt and mineral dust. The chemical transformations of the injected gaseous SO_2 into H_2SO_4 which then forms aerosols are performed by HAM.

Cirrus cloud formation is based on *Kärcher et al.,* [2006] and *Hendricks et al.,* [2011]. In order to allow supersaturation with respect to ice necessary for cirrus cloud formation, we got rid of the saturation adjustment scheme and solve the depositional growth equation. Orographic cirrus clouds which form in the lee of mountains due to gravity waves are parametrized following *Joos et al.,* [2008]. The vertical velocity is the sum of a large-scale component and a turbulent component, where the turbulent component either depends on the turbulent kinetic energy or on the gravity wave drag. In the altitude where cirrus clouds can form the large scale component of the vertical velocity is negligible and the turbulent component dominates.

Ice nucleation and depositional growth in cirrus clouds takes place in a competitive manner [*Kärcher et al.,* 2006; *Hendricks et al.,* 2011]: Homogeneous freezing of monodisperse solution droplets and heterogeneous freezing of internally as well as externally mixed dust aerosols compete for the available water vapor. Also, depositional growth of pre-existing ice crystals is taken into account and may suppress the formation of new ice crystals. Homogeneous freezing follows the description of *Koop et al.,* [2000]. Heterogeneous freezing is based on *Moehler et al.,* [2006, 2008]. Even though our simulations show that heterogeneous freezing takes place, it is hardly affected by GE. The general contribution of the heterogeneously formed ice crystals is small compared to homogeneously formed ones. Both the GE and the control simulation were run over five years starting from 1991 and ending in 1995 after a spin-up of one year. These years where chosen because of the eruption of Mt. Pinatubo in 1991. We used prescribed sea surface temperatures and sea ice distributions.

3. Results

Figure 1a shows the aerosol forcing represented as the change in the globally averaged number concentrations of solution droplets as function of time and pressure. GE leads to enhanced soluble aerosol concentrations in the lower stratosphere because the injected gaseous SO_2 reacts with OH and H_2O to form H_2SO_4 particles. Our simulations show that if there was a way of injecting SO_2 continuously, a rather stable aerosol layer in the stratosphere could be established by GE.

The major sink of stratospheric aerosols is sedimentation by gravitational settling, thus they enter the troposphere, where they are removed from the atmosphere within a few weeks.

Do these sedimenting aerosols have the potential to change the microphysical and radiative properties of cirrus clouds? This question is answered in Figure 1b depicting the change in ice crystal number between the geoengineered and undisturbed atmosphere. It is clearly shown that GE leads to cirrus clouds with fewer ice crystals compared to the reference simulation. Cirrus clouds mainly form between 100-400 hPa where the reductions in ice crystal number vary between 10-500 l^{-1}. Comparing that to the absolute values in the undisturbed atmosphere illustrates that the ice crystal number is reduced by 5-50% due to GE. Ice formation in cirrus clouds is mainly driven by homogeneous freezing [*Jensen et al.,* 1998]. Homogeneous freezing describes the formation of ice crystals by spontaneous freezing of solution droplets below temperatures of 238 K [*Koop et al.,* 2000]. It mainly depends on temperature and vertical velocity, whereas the aerosol size distribution plays only a minor role [*Kärcher and Lohmann,* 2002]. Higher vertical velocities cause higher supersaturation such that more ice crystals can nucleate before the supersaturation is depleted by depositional growth (=deposition of water vapor onto the ice crystals). For the temperature the dependence is inverse: Lower temperatures lead to more ice crystals because the lower the temperature the slower the depositional growth and the higher the nucleation rate. Figures 1c and d indicate that GE has an impact on the temperature and the vertical velocity in the upper troposphere and lower stratosphere (UTLS). The injected aerosols scatter solar radiation back to space, but also absorb and re-emit terrestrial radiation leading to a cooling of the Earth's surface and a warming of the UTLS. The eruption of Mt. Pinatubo led to a heating of the stratosphere of up to 3.5 K in some regions and cooled the Earth's surface globally by 0.5 K [*McCormick et al.,* 1995]. This change in vertical temperature distribution induces a stabilization of the troposphere, represented by the reduced vertical velocity (Figure 1d). Our simulations suggest that the enhanced temperature and reduced vertical velocity directly influence the rate of homogeneously formed ice crystals (not shown here). Thus GE leads to considerable reductions of the homogeneously formed ice crystals in regions of enhanced temperatures and decreased vertical velocity.

We compare the stratospheric heating simulated by our model versus temperature anomalies at a height of 30 hPa caused by the eruption of Mt. Pinatubo in June 1991 given by ERA-40 reanalysis data in Figure 2. The anomalies were derived by computing the difference between the Mt. Pinatubo period (June 1991 to May 1993) from the climatological mean, where the climatological mean was computed using monthly data over 43 years from January 1958 to December 2001 [*Thomas et al.,* 2009]. The eruption of Mt. Pinatubo was a single event where approximately 18 Mt of SO_2 were injected into the stratosphere within a few days [*McCormick et al.*, 1995]. The GE simulations on the other hand are based on continuous SO_2 injections of 5 Mt SO_2/a. The amount of injected SO_2 as well as the time-frame and the location of the injections is different between the two data sets. Thus, the stratospheric heating can only be compared in a qualitatively manner. ECHAM5-HAM shows similar stratospheric temperature anomaly patterns to those in ERA40 (Figure 2), with the exception of the poles where our model is not able to represent the seasonality of the stratospheric temperature anomalies correctly. Keeping the limitations of this comparison into account we conclude that ECHAM5-HAM represents the stratospheric heating due to stratospheric aerosols reasonably well.

Figure 3 shows the optical depth of cirrus clouds in a geoengineered and undisturbed atmosphere. We see that cirrus clouds in a geoengineered atmosphere become optically thinner due to a decreased ice crystal number concentration and mass. Optically thinner cirrus clouds lower the planetary albedo and more short-wave (SW) radiation enters the Earth-atmosphere system, leading to a warming. On the other hand, optically thinner cirrus clouds trap less long-wave (LW) radiation such that more terrestrial radiation is emitted to space, inducing a cooling. For moderate to thin cirrus clouds the LW effect dominates over the SW effect (because the emissivity changes stronger with optical depth than the reflectance, see *Gettelman et al.,* [2012])

Table 1 shows the changes in the radiative fluxes and cloud adjustments at the top of the atmosphere between the geoengineered and undisturbed atmosphere. The scattering of solar radiation by the injected aerosols under clear sky conditions is -1.49 W m^{-2}. The LW aerosol forcing is only slightly affected by the absorption and re-emission of infrared radiation. The net clear sky aerosol forcing of -1.53 W m^{-2} illustrates that the GE aerosols cool the Earth-atmosphere system in general. Assuming a climate feedback parameter of 1.08 W m^{-2} K^{-1} [*Andrews et al.,* 2012] shows that the injection of 5 Mt SO_2/a could counterbalance ~1.4°C of warming if the clear sky cooling would be the same as the all sky cooling. However, in the presence of clouds the radiative fluxes can be changed significantly [*Schulz et al.,* 2006]. *Stier et al.,* [2012] found a decrease in the all sky aerosol forcing compared to the clear sky aerosol forcing by a factor of two for scattering aerosols. Thus we can assume that the negative aerosol forcing under all sky conditions is significantly less than 1.49 W m^{-2}.

The cloud adjustments in the LW are -0.51 W m^{-2} and in the SW 1.1 W m^{-2}. As described above LW effects dominate over SW effects for thin cirrus clouds. Thus we can assume that the contribution of optically thinner cirrus to the SW cloud adjustment is less than |ΔLW cloud adjustment| (<-0.51 W m^{-2}) and that the majority of the SW cloud adjustment is caused by the mere presence of clouds as described by *Schulz et al.* [2006] and in accordance with *Stier et al.* [2012].

The net GE forcing exerted to the Earth-Atmosphere system for all sky conditions is -0.93 W m^{-2}. The main contributions (60%) are from LW effects which are clearly caused by the thinning of cirrus clouds. The SW contributions (40%) are affected by both the thinning of cirrus clouds and the presence of clouds.

4. Conclusions

We performed idealized simulations to study possible microphysical and dynamical impacts of stratospheric GE on cirrus clouds. We find that cirrus clouds become optically thinner in a geoengineered atmosphere due to a reduced homogeneous nucleation rate which is caused by a warming and stabilization of the UTLS. Our simulations suggest that GE in terms of stratospheric aerosols can cool the Earth-atmosphere system. Stratospheric injections of 5 Mt SO_2/a lead to an overall negative forcing of 0.93 W m^{-2} in our simulations balancing ~0.85°C of warming assuming a climate feedback parameter of 1.08 W m^{-2} K^{-1} [*Andrews et al.*, 2012]. This overall forcing to the Earth-Atmosphere system has contributions from the GE aerosols themselves and from modified cirrus clouds. We find that in the LW, optically thinner cirrus clouds lead to a strong cooling which contributes to the overall forcing by 60%. The SW cloud adjustments show a strong positive value, but cannot clearly be separated into a cirrus cloud forcing and a weakening of the aerosol forcing induced by the mere presence of clouds [*Schulz et al.*, 2006].

Our analysis of the impact of GE on cirrus clouds and the implications for GE effectiveness do not consider all factors. One shortcoming of this study is that we used prescribed sea surface temperatures. It will be important to redo the simulations with a mixed layer ocean (MLO) in order to allow the ocean to respond to the applied aerosol forcing. The difference between an atmospheric GCM and a GCM-MLO simulation could however be small [*Hansen et al.*, 2007].

Another shortcoming is that our model does not include ozone chemistry but uses prescribed ozone fields. Therefore, the temperature response to the stratospheric aerosols may be overestimated, because GE destroys the ozone layer partially [*Heckendorn et al.,* 2009; *Tilmes et al.,* 2008] while ozone itself heats the stratosphere. Ozone destruction would thus lead to a cooling which is not considered in our study. The stratospheric heating in our simulations is about 2 K. Observations after the eruption of Mt. Pinatubo show peak anomalies of 3.5 K [*McCormick et al.,* 1995] and also other modeling studies suggest stratospheric heating about 1-3 K depending on aerosol loading. More modeling studies with coupled ozone chemistry would be desirable to fully address the question of the implications of stratospheric GE.

In order to reproduce the effect of GE on cirrus clouds homogeneous freezing has to be parametrized depending on temperature, vertical velocity and aerosol size distribution as shown by several studies [*Koop et al.,* 2000; *Kärcher and Lohmann,* 2002]. Our results are different to *Lohmann et al.,* [2003] who parametrized homogeneous freezing depending on the above mentioned variables, but found only a weak impact of volcanic aerosols on cirrus clouds. The discrepancy can be explained by the aerosol scheme used: *Lohmann et al.,* [2003] prescribed the injected aerosol distributions in ECHAM which were calculated previously by an external aerosol model. Thus, they were not able to include the stratospheric heating caused by the absorption of infrared radiation. This further implies that GE studies mimicking the effect of stratospheric aerosols by reducing the solar constant are not sufficiently good analogs because also they lack the indirect aerosol effects, e. g. the stratospheric heating and thus indirect effects on cirrus clouds.

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Figure captions:

Fig. 1 The temporal evolution of global monthly mean changes between the geoengineered and undisturbed atmosphere of the following variables: soluble aerosol number concentration, total ice crystal number concentration (*ICNC*), temperature (*T*) and vertical velocity (*w*). The black solid line in the upper left panel indicates the global monthly mean tropopause height.

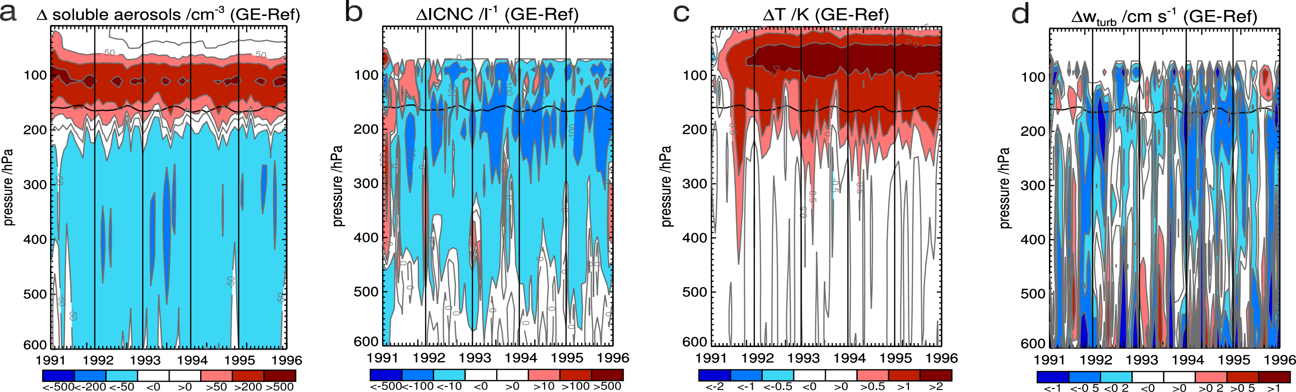
Fig 2 Temporal evolution of monthly and zonally averaged temperature anomalies at 30 hPa for the time period of June 1991 to May 1993. In the left panel temperature anomalies caused by the eruption of Mt. Pinatubo in June 1991 of ERA-40 reanalysis data are shown. In the right panel the change in temperature between the geoengineered and undisturbed atmosphere simulated by ECHAM5-HAM is shown.

Fig 3 Zonal mean cirrus optical depth in the geoengineered and undisturbed atmosphere averaged over the years 1991-1995 .

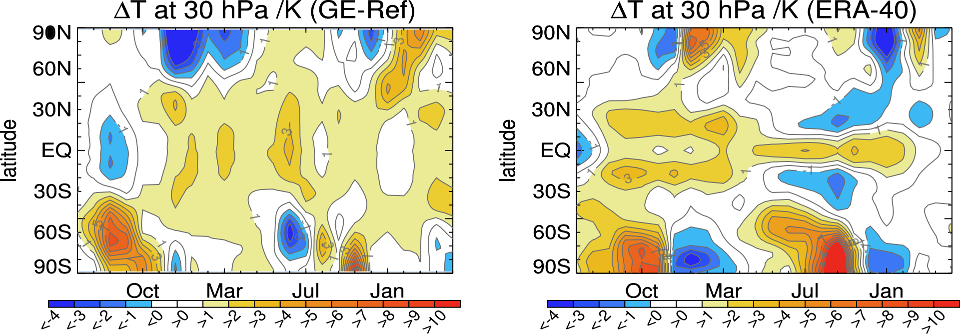
Tables:

	ΔSW /W m^{-2} (GE-Ref)	ΔLW /W m^{-2} (GE-Ref)	ΔNet /W m^{-2} (GE-Ref)
Clear sky	-1.49	-0.05	-1.53
Cloud adjustment	1.11	-0.51	0.6
All sky	-0.37	-0.56	-0.93

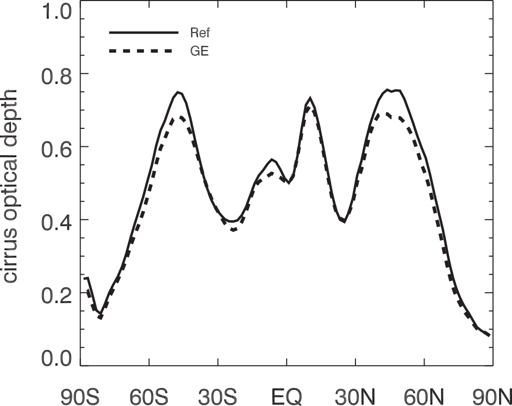
Table 1) Global mean changes in the radiative fluxes and cloud adjustment at the top of the atmosphere between the geoengineered and undisturbed atmosphere averaged over the years 1991-1995.



a
Δ soluble aerosols /cm-3 (GE-Ref)
b
ΔICNC /l-1 (GE-Ref)
c
ΔT /K (GE-Ref)
d
Δw_turb /cm s-1 (GE-Ref)
pressure /hPa
100
200
300
400
500
600
1991
1992
1993
1994
1995
1996
<-500 <-200 <-50 <0 >0 >50 >200 >500
<-500 <-100 <-10 <0 >0 >10 >100 >500
<-2 <-1 <-0.5 <0 >0 >0.5 >1 >2
<-1 <-0.5 <0.2 <0 >0 >0.2 >0.5 >1



ΔT at 30 hPa /K (GE-Ref)
ΔT at 30 hPa /K (ERA-40)
latitude
90N
60N
30N
EQ
30S
60S
90S
Oct
Mar
Jul
Jan
<-4 <-3 <-2 <-1 <0 >0 >1 >2 >3 >4 >5 >6 >7 >8 >9 >10



Ref
GE
cirrus optical depth
0.0
0.2
0.4
0.6
0.8
1.0
90S
60S
30S
EQ
30N
60N
90N