The dependency of geoengineered sulfate aerosol on the emission strategy

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Abstract
The climatic effect of geoengineered stratospheric sulfate aerosol depends on the strategy for sulfur emission that determines the microphysical evolution of the resulting sulfate layer, in particular the radius and radiative impact of the aerosols. Simulations with a three-dimensional general circulation model (GCM), including an aerosol microphysical model, show e.g. decreasing sulfate lifetime with increasing emission rate. Furthermore, scenarios that differ with respect to location, local and temporal distribution and chemical composition of the emissions are studied. The study shows detailed information on particle radius, sulfate burden and radiative impact for these scenarios. Copyright © 2010 Royal Meteorological Society

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1. Introduction

There is increasing scientific evidence that the anthropogenic emission of greenhouse gases has a significant impact on the Earth’s climate (IPCC, 2007). Despite attempts to limit anthropogenic emissions, carbon dioxide concentrations in the atmosphere have continued to increase. Consequently, a debate has started on technical methods to limit global warming, usually referred to as ‘geoengineering’. One of the proposed techniques, suggested by Budyko (1977) and Crutzen (2006), follows the natural example of volcanic eruptions emitting large amounts of sulfur dioxide (SO2) into the stratosphere. Chemical and microphysical reactions cause the formation of sulfate aerosols that reduce the incoming solar radiation. It is however unclear, as to what extent this volcanic example can be used as analogue for the effect of geoengineered aerosol because of possible differences in the aerosol size distributions and radiative properties resulting from different emission patterns.

Recently, several studies have estimated the climatic effect of geoengineered aerosols by performing climate model simulations with prescribed particle size and relatively coarse assumptions for the particle evolution (Rasch et al., 2008; Robock et al., 2008; Tilmes et al., 2009). The first study with a more comprehensive, albeit two-dimensional, sectional aerosol microphysical model has been performed by Heckendorn et al. (2009). One important result of this study is that the particle size distribution of the aerosol cloud depends strongly on the magnitude of the injections. Our study can be seen as an extension of the one by Heckendorn et al. (2009) using a three-dimensional modal aerosol model and allowing for dynamical feedbacks on the particle distribution. The particle size is a crucial parameter for the effectiveness of stratospheric aerosols as it influences its absorption and scattering properties. For the temporal development of the aerosol size distribution, the injection strategy is likely of importance. Sulfur could be injected at different rates and different heights, continuously or pulsed, in a single small area or distributed along the Equator. To study the impact of different emission strategies on the evolution of the sulfate aerosol and in particular the resulting particle radius and radiative impact, we have performed simulations with the middle atmosphere version of the general circulation model (GCM) ECHAM5 (Roeckner et al., 2006) interactively coupled to a modified version of the aerosol microphysical model, HAM (Stier et al., 2005).

It is assumed that one significant side effect from sulfate geoengineering is additional stratospheric ozone depletion. In contrast to the work of Heckendorn et al. (2009), our model does not include stratospheric ozone chemistry. However, we analyze the surface area density (SAD) of geoengineered aerosols as an indicator for its ozone depletion potential (Tilmes et al., 2008).

2. Model description and setup of simulations

The simulations were performed with the middle atmosphere model MAECHAM5 (Giorgetta et al., 2006) in a version with spectral truncation at wavenumber 42 (T42) and 39 vertical layers up to 0.01 hPa. The GCM solves prognostic equations for vorticity, divergence, surface pressure and temperature. The aerosol microphysical model HAM was interactively coupled to the GCM. HAM calculates the formation of sulfate aerosol which includes nucleation, accumulation,
condensation and coagulation processes. M7 (Vignati et al., 2004), the microphysical core of HAM, was modified to allow for a better representation of stratospheric sulfur aerosol according to box-model studies for large volcanic eruptions (Kokkola et al., 2009). The changes include a reduction to three modes (nucleation, Aitken and accumulation), and a smaller standard deviation of the accumulation mode (σ = 1.2 instead of 1.59). Details on sulfur chemistry and prescribed gaseous species concentrations are described by Niemeier et al. (2009). The simulations include only sulfate aerosol. Besides the geoengineered sulfur, only dimethyl-sulfide (DMS) and carbonyl-sulfide emissions are included in the simulations.

The model setup has been used in previous studies on the climatic impact of volcanic eruptions and compared in detail with measurements after the Mount Pinatubo eruption (Niemeier et al., 2009). The results show good agreement with measurements. We see a slight overestimation of the poleward transport, simulated particle radius, and aerosol optical depth (AOD).

To study the dependence of the particle size distribution and the effects of geoengineered sulfate aerosol on the amount of injected SO2, a series of numerical experiments were designed with continuous emissions of 1, 2, 4, and 8 Mt(S)/y. In most experiments, sulfate was injected into one grid box with the size of 2.8° × 2.8° centered at 1.4° N 121° E, a similar position as used by Robock et al. (2008). We conducted experiments with emissions at 60 hPa (~19 km) height (Exp60) and 30 hPa (~25 km) height (Exp30). The lower injection level, 60 hPa, can still be reached by some planes. For the higher level, 30 hPa, a higher lifetime of aerosols is expected. Robock et al. (2009) show options to release particles even at such high stratospheric altitudes. Three further sensitivity experiments were performed for the 4 Mt(S)/y emission scenario: SO2 was emitted twice a year in pulses of 30 days each (Epulse) into the box given above and emitted in one latitude band (Elat) of the width of one grid box, along the Equator. In experiment (Eh2so4), emissions into one grid box are assumed to be in the form of H2SO4 instead of the precursor gas SO2, as suggested by Pierce et al. (2010). All presented results are averaged over 3 years of a steady-state sulfate layer.

3. Results

3.1. Role of emission rate

The simulated sulfate load depends strongly on the emission rate (Figure 1). For emissions at 60 hPa (Exp60), maximum concentration and global burden increase by a factor of 1.8–1.9, when doubling SO2 injection. Accordingly, the lifetime decreases with increasing emission strength. For emission rates of 1, 2, 4 and 8 Mt(S)/y, we obtain lifetimes of 0.91, 0.85, 0.80 and 0.74 years, respectively. The size of the particles increases with increasing sulfate load (Figure 2(b)): from an effective radius of 0.26 µm (1 Mt(S)/y) to 0.4 µm (8 Mt(S)/y). The calculated burden and its dependency on the emission rate are similar to the values obtained by Heckendorn et al. (2009). However, their global burden (Figure 2(a)) increases less strongly with the emission rate, possibly due to differences in the particle sizes resulting from the use of different aerosol models (sectional vs modal). Rasch et al. (2008) show global burdens for emission rates of 1 and 2 Mt(S)/y, which are about a factor of 2 larger than our values. This is likely due to their assumption of fixed, uniform radii, which prevent the growth of large, fast sedimenting particles.

The annually averaged aerosol distributions shown in Figure 1 result in an almost zonal distribution of AOD (not shown). The AOD is also characterized, besides the increase with increasing injection rate, by rather symmetric meridional distributions with secondary maxima in the mid-latitudes (Figure 3, bottom). This pattern is mirrored by the resulting aerosol radiative forcing shown as the annual mean clear-sky shortwave (SW)-flux anomaly in Figure 3 (top). Flux anomalies are computed in the model via a twofold calculation of the radiation, with and without aerosols, at each radiation time step. The pronounced mid-latitude maxima are mainly summertime effects. The low values at the poles are the impact of seasonal solar flux conditions. Although during two Summer months the values close to the poles are in the range of tropical values, they are zero between late Autumn and early Spring resulting in low annual mean values. SW-flux (all-sky) anomalies show a strong single maximum in the Tropics. Differences in the clear-sky forcing are related to cloud albedo effects.

Meridional transport in the stratosphere is caused by the Brewer–Dobson circulation (BDC). Apparently, the strength of the BDC differs between the models used for geoengineering studies, causing different results for AOD and SW-fluxes. Heckendorn et al. (2009) show similar clear-sky SW-flux maximum values, but much lower ones in the mid-latitudes, which is likely due to very weak meridional transport. Pattern and values of downward SW-flux of Robock et al. (2008), in a study with fixed aerosol size, are comparable to our results in mid-latitudes. Similar to MAECHAM5, they indicate a slightly stronger meridional transport compared to measurements after the Mount Pinatubo eruption. These differences are reflected in the global SW radiative forcing (Figure 2(c)). SAD increases with increasing emissions and are slightly stronger at high latitudes than in the Tropics (Figure 4). Values in Heckendorn et al. (2009) are similar in the tropical maxima, but much lower poleward.

3.2. Role of emission height

The cases with emissions at 30 hPa (Exp30) shows, compared with the same emission rates at 60 hPa,
higher concentrations (Figure 2(a)) and about 10–20\% larger particle radii (Figure 2(b)). A longer sedimentation path and slightly less poleward transport, indicated by more pronounced maxima of AOD and SW-flux in the equatorial region (Figure 3, right), contributes to a longer aerosol lifetime in the stratosphere. The global burden is increased by a factor of 1.7, 1.6, 1.5 and 1.37 for emissions of 1, 2, 4 and 8 Mt(S)/y, respectively, compared to the 60 hPa case. The 8 Mt(S)/y rate yields particles similar in effective radius (10–20\% smaller) compared to measurements taken after the Mount Pinatubo eruption (Ansmann et al., 1997). For lower emission rates, the particles have an even smaller size (0.25–0.4 µm), confirming assumptions of Crutzen (2006).

The SAD is higher in the 60 hPa emission case (Figure 2(d)) caused by a higher number of fine nucleation particles (about ten times) in the Tropics. The relation between SAD and all-sky SW-flux anomaly highlights the higher ratio of effectiveness (radiative forcing) to risk (ozone depletion) of emissions at higher levels.

3.3. Role of emission period

To study the impact of the emission period we performed a sensitivity study with pulsed SO$_2$ emissions equivalent to 4 Mt(S)/y at 60 hPa (Epulse). Results are very similar compared to continuous emissions. Yearly averaged global sulfate burden and the SW-fluxes are almost identical (Figure 2, red circle). Particle size is 0.02 µm smaller.

3.4. Role of emission area

Simulation Elat with SO$_2$ emission homogeneously distributed along the Equator shows a different picture.
compared to the simulations with grid-box emissions. The particle size increases to 0.4 μm (Figure 2(b), red asterisks), causing stronger sedimentation and lower sulfate concentrations, lower AOD and up to 0.4 W/m² lower SW-fluxes (all-sky) (Figure 3, light red lines).

Emitted into one grid box, SO2 and H2SO4 values are larger between 0 and 120° E than farther east of the emission area. The longitudinal emissions cause a smaller SO2 emission flux and equally distribute H2SO4 in low concentrations, which lead to condensation on existing particles rather than to nucleation, causing low particle numbers with an increased radius. Therefore, a global spread of the injection band seems to be contra productive for geoengineering purposes. This result adds the third dimension to the results of Heckendorn et al. (2009), where emissions had to be assumed as zonal homogeneous.

3.5. Role of emitted species

The opposite effect shows simulation Eh2so4, where sulfur is assumed to be emitted in the form of H2SO4 rather than SO2. The high initial H2SO4 concentration favours nucleation over condensation, causing a high amount of small particles, less effective coagulation and therefore more but smaller particles in the accumulation mode (0.23 μm). Therefore, the lifetime is increased by 1.5 months and radiative forcing is up to 20% stronger in the Tropics (Figure 3, dark red lines). The AOD increased mainly in the Tropics.

3.6. Role of dynamical feedback

As described by Niemeier et al. (2009) for the case of volcanic eruptions, the additional heating due to absorption by sulfate aerosol may lead to increased upwelling. This should lead to an increased aerosol lifetime. To quantify this feedback and the possible effect on the radiative forcing, an additional experiment (with 4 Mt(S)/y emitted at 60 hPa) was performed with the stratospheric aerosol heating switched off.

Figure 5 shows an increase in the sulfate mass-mixing ratio up to 1 ppb (~20%) in the upper part of the sulfate layer and a slight decrease in the lower part caused by the dynamical feedback when the aerosol heating is taken into account. The residual (upward) vertical velocity in the equatorial stratosphere is increased by about 5–10% as a result of the radiative heating.

4. Conclusions

Sulfur emission into the stratosphere has been suggested as a geoengineering option to cool the Earth. The presented model study tries to estimate the efficiency of different emission strategies by studying the microphysical behaviour of the resulting aerosol. Our numerical experiments confirm an important earlier result obtained by Heckendorn et al. (2009). An increase in the SO2 emission rate does not lead to a similar increase in the radiative forcing because
Figure 3. Annual and zonal mean of SW-flux anomaly (W m⁻²) at the surface for clear-sky (top) and all-sky (with clouds) conditions (middle) and aerosol optical depth (bottom). Contours are given for emission rates of 1 (orange), 2 (blue), 4 (red) and 8 Mt(S)/y (gray), simulation Elat (red cross) and simulation Eh2so4 (dark red). All results are given for emissions at 60 hPa (left) and 30 hPa (right).

Figure 4. SAD (mm²/m³) for SO₂ emissions at 60 hPa.

the size of the aerosols increases and, accordingly, its lifetime decreases. Studies that use fixed aerosol parameters independent of the emission rate may therefore overestimate the radiative forcing at high emission rates.

Further, our study shows that the lifetime of the particles depends on emission height. The calculated burden for emissions at 30 hPa is almost twice the burden calculated for the same emission strength but at 60 hPa, resulting in a significantly larger forcing despite the growth of larger particles. A higher injection level results in a smaller SAD but the maximum SAD is located at a similar height as the maximum of the ozone concentration. Studies with included chemistry models may show whether the ozone-depletion changes with changing emission height. However, emission at 30 hPa would probably be technically more challenging than at 60 hPa. Direct emissions of H₂SO₄ instead of SO₂ decreases the availability of H₂SO₄ outside the emission region and increases the lifetime of the sulfate by about 15%. Because of using a three-dimensional aerosol model, we could compare the effect of emissions in one single grid box versus emissions along the Equator. The second option causes much larger radii, shorter lifetime and lower radiative impact. However, it should be noted that results of our and other previous studies may suffer from neglecting the microphysical process taking place on small spatial and temporal scales during the injection. The use of an interactively coupled aerosol and general
circulation model has allowed us to study the dynamical feedback on the aerosol cloud resulting from the additional heating. According to our results, this effect is significant but small. Hence, one can assume that the error made in earlier studies through the use of non-interactive coupling is relatively small.

Some additional model deficiencies might influence our results. We have used a coarse grid version of MAECHAM5 that does not simulate a quasi-biennial oscillation (QBO). Hence, the impact of different QBO phases on the transport of the aerosol cloud (Punge et al., 2009) is not taken into account. As our model neglects the stratospheric ozone chemistry, the effect of possible ozone changes on radiation are also missing.

Our study shows that radiative forcing of geoengineered sulfate aerosols depends strongly on the emission strategy. The design of our study, however, does not allow an assessment of possibly important side effects of this geoengineering technique. To study, in particular, the associated ozone depletion and a possible change in precipitation patterns, long ensemble simulations with climate models incorporating a comprehensive representation of chemistry and aerosol microphysical processes would be needed. A scientific consensus on these side effects is still to be reached and very likely requires further fundamental progress in climate research.

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