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Sulfate geoengineering: a review of the factors controlling the needed injection of sulfur dioxide

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Abstract.

Sulfate geoengineering has been proposed as an affordable and climate-effective means for temporarily offset the warming produced by the increase of well mixed greenhouse gases (WMGHG). This climate engineering technique has been planned for a timeframe of a few decades needed to implement global inter-governmental measures needed to achieve stabilization of the atmospheric content of WMGHGs (CO_2 in particular). The direct radiative effects of sulfur injection in the tropical lower stratosphere can be summarized as increasing shortwave scattering with consequent tropospheric cooling and increasing long-wave absorption with stratospheric warming. Indirect radiative effects are related to induced changes in the ozone distribution, stratospheric water vapor abundance, formation and size of upper tropospheric cirrus ice particles and lifetime of longlived species, namely CH_4 in connection with OH changes through several photochemical mechanisms. A direct comparison of the net effects of WMGHG increase with direct and indirect effects of sulfate geoengineering may help fine-tune the best amount of sulfate to be injected in an eventual realization of the experiment. However, we need to take into account large uncertainties in the estimate of some of these aerosol effects, such as cirrus ice particle size modifications.

1 Introduction

The overwhelming evidence of a warming of surface temperatures on Earth caused by the anthropogenic increase in greenhouse gases (GHG) has forced the scientific community to look for methods of mitigating and possibly reversing this trend (Solomon et al. (2007)). Such a need is made even more pressing if we look at the projections for the next century. The International Panel for Climate Change (IPCC) has built various Representative Concentration Pathways (RCPs) predicting future anthropogenic emissions (greenhouse gases, anthropogenic aerosols, short lived gas species etc.) and assessed the effect of such scenarios on the Earth's climate using a series of multi-model experiments (CMIP5) (Taylor et al. (2012)). The main result is the agreement among most models on a warming of the Earth's surface ranging from a 1 K increase by 2100 for the most optimistic scenario (RCP2.6, with near-constant emissions between 2020 and 2100) to a 3.7 K increase for the least optimistic scenario (RCP8.5, with most developing countries increasing their emissions sensibly) (Meinshausen et al. (2011)). These forecasts tell us that,

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even with the most optimistic emission scenario, a sudden reversing of the temperature trend is not expected (Solomon et al. (2007);Nordhaus (2007)).

In order to mitigate the effects that such a warming would have on the climate of our planet, some methods have been proposed to balance out the direct effects of GHG, generally known under the name of climate engineering or geoengineering. Geoengineering methods have to be carefully evaluated on four grounds: effectiveness (the potential for the proposed method to work), affordability, timeliness (how long it would take to deploy it and how fast would it work) and safety (the risks linked with the deployment of the method). Such geoengineering methods would hopefully need to be applied only during the so called transition period (2020-2070), between fossil and clean energy sources (Kravitz et al. (2011)). These methods can be divided into two large groups: the first group is composed of carbon dioxide removal techniques, whose aim is to directly reduce the amount of carbon dioxide in the atmosphere by means such as afforestation, atmospheric CO_2 scrubbers, in-situ carbonation of silicate over land, and fertilization and alkalinity enhancements over the oceans. The second group, in which the method we will be studying further on is situated, is the one known under the term Solar Radiation Management techniques, whose aim is to decrease the amount of incoming radiation on the Earth surface: among those we find surface albedo increase, cloud albedo enhancement, space-based reflectors, and stratospheric aerosol injection, also called sulfate engineering (CEC (2014)).

Sulfate geoengineering (SG) prescribes the sustained injection of sulfur dioxide (SO_2) in the tropical lower stratosphere, originally proposed by Budyko (2013) and further developed by Crutzen (2006). Under the international modeling project GeoMIP (Geoengineering Model Intercomparison Project; Robock et al. (2011); Kravitz et al. (2012); Kravitz et al. (2013)). chemistry-climate models and atmosphere-ocean coupled models have been used to explore the radiative, chemical and dynamical modification of climate by SO_2 injection. Several studies were conducted to compare a control simulation ensemble under the IPCC scenario RCP4.5 (Taylor et al., 2012) [2] and a sulfate geoengineering simulation. In this review we summarize the climate effects of a constant stratospheric injection of SO_2, such as the one prescribed by the GeoMIP experiment G4, where 5 Tg/year of SO_2 were injected in the tropical lower stratosphere from 2020 to 2070 (Pitari et al. (2014a) Aquila et al. (2014)), rather than a time-varying injection, such as in the GeoMIP experiment Robock et al. (2011), where the amount of the injected SO_2 changes year-by-year in order to keep the top-of-atmosphere (TOA) radiative balance constant. This is because the G4 approach (even if with different amounts of constant SO_2 injection) has been used and documented in a wider number of studies (see also Heckendorn et al. (2009); English et al. (2012); Niemeier and Timmreck (2015)).

The direct effect of an injection of SO_2 is an increase in the local concentration of optically active H_2O-H_2SO_4 aerosol particles in the lower stratosphere. These particles increase the amount of back-scattered solar radiation, resulting in less radiation arriving at the Earth's surface, thus cooling the whole troposphere. The idea itself of sulfate geoengineering comes from the observation of various explosive volcanic eruptions over the last century, which injected large amounts of sulfur in the lower stratosphere over a very short amount of time and whose direct impact on the global mean surface temperature has been known for some time (Robock and Mao (1995)).

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2 Review of radiative forcing effects

2.1 Direct forcing of stratospheric sulfate

The underlying physical processes behind the injection of SO_2 into the atmosphere have been widely studied thanks to the various explosive volcanic eruption of the 20^{th} century. For instance, after the Mount Pinatubo eruption of June 1991, where 7 to 10 Tg-S where injected into the stratosphere (Read et al. (1993); Krueger et al. (1995)), a significant drop in surface temperature of about 0.5 K was observed in the year following the eruption (Dutton and Christy (1992)), along with a sharp reduction in the TOA net radiative flux (3 to 10 W/m^2) right after the eruption (Minnis et al. (1993)). These effects can be explained by SO_2 oxidation into SO_4 followed by the formation of H_2O-H_2SO_4 supercooled liquid droplets, which create an optically active thick cloud that reflects part of the incoming solar radiation. This results in a surface cooling and a local stratospheric warming. The stratospheric warming is due to changes in diabatic heating rates produced by aerosol absorption of solar near infrared and planetary radiation and by the ozone absorption of the additional UV radiation scattered by the volcanic aerosols (Pitari (1993)).

When considering the effects of the proposed injection of sulfur into the atmosphere, however, a series of factor must be taken into account, complicating the analogy between these kind of geoengineering experiments and volcanic eruptions. Obviously, the amount of sulfur and the height and latitude at which it is injected in a geoengineering experiment all play a prominent role in its related effects. Some recent papers, such as English et al. (2012) and Niemeier and Timmreck (2015) analyzed a series of different geoengineering experiments accounting for the different factors previously mentioned. Their results show that the relation between injected SO_2 and the sulfate mass burden is non-linear, with larger injection rates producing a lower efficiency of SG. This is due to the fact that injections of larger amounts of SO_2 lead to the formation of larger aerosol particles by gas condensation, which are rapidly removed from the stratosphere by gravitational settling. Aside from the reduction in the aerosol lifetime, the size of the produced aerosol particles also influences the amount of scattered radiation, because the sulfate scattering efficiency peaks at a particle radius of around 140 nm and decreases as aerosols become larger. The highest burden to injection ratio is achieved for stratospheric injections between 30N and 30S (English et al. (2012)). The altitude also plays a significant role in determining the aerosol lifetime, due to a faster sedimentation removal in the upper troposphere when the sulfur injection is localized closer to the tropical tropopause layer (TTL) (Aquila et al. (2014)).

As shown in Pitari et al. (2014a), the injection of 5 Tg-SO_2/yr produces, according to the models used in the experiment G4, a net radiative forcing (RF) between -1.54 W/m^2 and -0.73 W/m^2. The different results are mainly dependent on the (calculated, or imposed in one case) different aerosol optical depth (AOD) and size distribution among models. However, while on the one hand Pitari et al. (2014a) show that SG leads to the desired effect of offsetting the positive RF of increasing well mixed greenhouse gases (WMGHG), on the other hand they show that SG side effects such as lower stratospheric warming must be carefully studied.

Enhanced lower stratospheric diabatic heating rates after major explosive volcanic eruptions and the consequent temperature increase were well documented both in observations (Labitzke and McCormick (1992); McLandress et al. (2015)) and through modeling experiments (Aquila et al. (2013); Pitari et al. (2016a)). The tropical lower stratospheric warming induces a signif-

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icant increase of westerly winds from the thermal wind equation, with peaks at mid-latitudes in the mid-stratosphere. These dynamical changes tend to increase the amplitude of planetary waves in the stratosphere and to enhance the tropical upwelling in the rising branch of the Brewer Dobson circulation (Pitari et al. (2014a)).

One of the possible consequences of such an enhancement is the modification of the quasi-biennial oscillation (QBO). The effects of the aerosol heating rates on the QBO under geoengineering conditions have been analyzed in the aforementioned study by Aquila et al. (2014) using the NASA Goddard climate-chemistry coupled model (GEOSCCM), which includes an internally generated QBO. Four different experiments were designed, using 5 Tg-SO_2/yr for the first two and 2.5 Tg-SO_2/yr for the others, injected at different altitudes (16-25 km and 22-25 km). They found that SG perturbs the QBO phase by prolonging the westerly phase in the 20-50 hPa layer proportionally to the stratospheric SO4 mass burden in the experiment (ranging from 1.5 Tg-S for the 16-25 km injection of 2.5 Tg-SO_2/yr to 4.7 Tg-S for the 22-25 km injection of 5 Tg-SO_2/yr).

Niemeier and Timmreck (2015) also mention a perturbation of the QBO in SG simulations performed with the ECHAM-HAM model. Their simulation includes explicit aerosol microphysics, so that the effects of the perturbed QBO on the aerosol size distribution are taken into account. They found that an injection of about 8Tg-S/yr would cause a slowing of the QBO oscillation with a constant QBO westerly phase in the lower stratosphere with overlaying easterlies, consistently with the findings by Aquila et al. (2014). The overall conclusion of both these studies is that a stratospheric sulfur injection could dramatically alter the QBO periodicity, up to producing a permanent westerly phase in the lower stratosphere, thus reducing the meridional transport efficiency (Trepte and Hitchman (1992)).

The SO_4 stratospheric lifetime in the simulations included in Aquila et al. (2014) was approximately 1.2 and 1.8 years for sulfur injection in the altitude layers 16-25 km and 22-25 km, respectively. However, it is interesting to note that the sulfate lifetime is systematically longer in the 5 Tg-SO_2/yr case with respect to the 2.5 Tg-SO_2/yr injection case ($\sim$1.9 years versus $\sim$1.7 years with injection in the 22-25 km layer and $\sim$1.25 years versus $\sim$1.2 years with injection in the 16-25 km layer). The higher heating rates produced by the aerosol in the 5Tg-SO_2/yr case are responsible for a stronger modification of the stratospheric circulation, resulting in the QBO changes and increased tropical upwelling, hence a better confinement of the particles in the tropical pipe (Trepte and Hitchman (1992); Pitari et al. (2016a)). This reduces the amount of aerosol that may be transported downwards across the extra-tropical tropopause in the lower branch of the BDC.

The prolonging of the aerosol lifetime found by Aquila et al. (2014), however, could be canceled if the microphysical effects of the QBO-dependent sulfur confinement in the tropical pipe were taken into account. In the simulations by Niemeier and Timmreck (2015) using the ECHAM-HAM model, which includes a representation of aerosol microphysics, the enhanced aerosol tropical confinement under conditions of a locked QBO westerly phase in the lower stratosphere decreases the SG aerosol lifetime, because the tighter tropical confinement of the aerosol also leads to larger particles and therefore a more efficient gravitational settling (U. Niemeier, personal communications).

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2.2 Indirect radiative forcing

In the following subsections we shall summarize the indirect changes caused by the SG-induced stratospheric warming and surface cooling. This section answers the question if any of these indirect effects could significantly counteract or enhance the primary goal of sulfate geoengineering of counteracting the positive RF from WMGHGs.

2.2.1 Ozone

Model simulations in Pitari et al. (2014a) showed that SG produces changes in stratospheric ozone due to a series of concurring factors, i.e., perturbation of photolysis rates because of the increased AOD, enhanced heterogeneous chemistry, and modifications of atmospheric dynamics. The models used in the G4 experiment showed significant changes in the ozone profile, with a decrease in the tropical column between 100 and 50 hPa mainly caused by a decreased O_2 photolysis, and a peak depletion at 30 hPa in the tropics for the combined effects of enhanced upwelling and losses in the chemical cycles. Above that layer, ozone was found to increase because of the reduction of NO_x via enhanced heterogeneous chemistry. Combined with similar changes in the extratropics, which are largely produced by modifications in the chemical processes, a total change in the global mean ozone column from -1.1 to -2.1 DU is calculated for the 2040-2049 decade. In terms of RF this produces a rather small negative result, ranging from -0.028 to -0.036 W/m^2.
After 2060, however, an ozone increase was predicted by the models, due to the decreasing amount of chlorine and bromine loading species, thus increasing the relative weight of the NOx catalytic cycle with respect to the others. The NO_x concentration, in turn, is decreased by heterogeneous chemical reactions on SG aerosols, so that ozone may globally increase.

2.2.2 Stratospheric water vapor

SG is expected to increase stratospheric water vapor concentrations by warming the TTL temperature. In the stratosphere, the water vapor concentration is regulated by the TTL temperature (Dessler et al. (2013)), combined with methane oxidation. The warmer the TTL temperatures, the more water vapor is able to enter the stratosphere. However, when considering the behavior of the TTL in a geoengineering scenario, we must consider two overlapping effects: an upper tropospheric cooling caused by the aerosol scattering, which cools the surface and stabilizes the troposphere (thus reducing convective heating), and a lower stratospheric warming caused by the infrared absorption by the aerosol particles. The amount of water vapor predicted in the stratosphere will thus depend on how the models represent these processes (Oman et al. (2008)).
Water vapor contributes to global warming, since it works as a GHG both in the troposphere and in the stratosphere (Forster F. and Shine (1999); Dessler et al. (2013)). Following the definition of radiative forcing, i.e., the net radiative flux change at the tropopause with fixed tropospheric temperatures and adjusted stratospheric temperatures, only stratospheric water vapor changes concur to the determination of the RF associated to any considered anthropogenic perturbation, SG in the present case. Pitari et al. (2014a) gave an estimate of the RF of the SG-induced increase in stratospheric water vapor. At 100 hPa in the tropics, 3 out of 4 models produce a warming ranging from +0.16 K to +0.58 K that leads to an increase in water vapor mixing ratio from 0.02 to 0.35 ppmv. This in turn produces a net positive RF ranging between +0.004 and +0.077 W/m^2. The fourth

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model, on the other hand, predicts a TTL cooling with a decreased amount of stratospheric H_2O and then a negative RF. This is partly due to an underestimated lower stratospheric aerosol warming, originated by an insufficient tropical confinement of the aerosol cloud.

2.2.3 Upper stratospheric ice

Several studies have proposed mechanisms by which the SG would affect upper tropospheric cirrus clouds, reaching, however, contradictory conclusions. Cirisan et al. (2013) found that SG directly provides ice nuclei (IN) of a larger size with respect to those in the unperturbed atmosphere, resulting in a rather small increase in cirrus cloud coverage. Kuebbeler et al. (2012), on the other hand, found that SG would decrease cirrus cloud coverage because of changes in temperature, vertical velocity and water vapor produced in the troposphere by the aerosol cooling effect. The aerosol driven surface cooling, coupled with the lower stratospheric warming, stabilizes the atmosphere due to a decreased vertical temperature gradient, thus reducing the available turbulent kinetic energy and the vertical updraft (Karcher and Lohmann (2002); Lohmann and Karcher (2002)). This results in a decrease of the upper tropospheric ice crystals formation, which in turn produces a less efficient trapping of the planetary longwave radiation and a reduction of the net atmospheric greenhouse effect.

As clearly demonstrated in a number of papers focusing on the physical processes taking place in the upper troposphere (Karcher and Lohmann (2002); Hendricks et al. (2011)), the formation of ice particles may take place via heterogeneous and homogeneous freezing mechanisms. Airborne measurements by Strom et al. (1997) reported typical concentrations of newly formed ice crystals of the order of 0.3 cm^{-3} in a young cirrus cloud at T=220 K in the upper troposphere of Northern Hemisphere mid-latitudes, in agreement with the model estimate of Karcher and Lohmann (2002) based on the assumption of ice particle formation via homogeneous freezing.

The homogeneous freezing mechanism normally dominates in the upper troposphere and involves water vapor freezing over liquid supercooled particles (as sulfate aerosols or sulfate coated aerosols), when the ice supersaturation ratio exceeds $\sim$1.5. In a SG perturbed atmosphere, more sulfate aerosols are available in the upper troposphere with respect to unperturbed background conditions thanks to extratropical downwelling and gravitational settling from the lower stratosphere. However, the background number density of sulfate aerosols in the upper troposphere is normally already much larger than the number of ice particles that can form (Karcher and Lohmann (2002)). This means that the SG driven increase of IN number density has basically no effect on the population of ice particles, but we may expect some impact on the ice particle size due to the larger size of IN made available by SG. This is the main conclusion of Cirisan et al. (2013), who note that the more large geoengineered particles exist (of typical sizes close to 0.5 μm), the less particles have to struggle against the Kelvin effect and more droplets may grow to larger sizes. This study analyzes in detail the direct SG impact on IN, as a complementary effect with respect to the dynamical indirect effect investigated by Kuebbeler et al. (2012). The main conclusion of Cirisan et al. (2013) is that the microphysical impact on cirrus clouds from geoengineered stratospheric sulfate aerosols is not an important side effect. They estimate a resulting mid-latitude average RF in the range of +0.02 W/m^2 to -0.04 W/m^2, depending on upwelling velocities and geoengineering scenario.This is consistent with the conclusions by Karcher and Lohmann (2002), who found

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that the effect of a perturbed aerosol size distribution on the ice particle population formed via homogeneous freezing is of secondary importance.

The other possible pathway for ice crystal formation is through heterogeneous freezing, which requires solid nuclei as mineral dust or freshly emitted black carbon. In this case, when the ice supersaturation ratio exceeds approximately 1.1, heterogeneous freezing may start Hendricks et al. (2011); sulfate aerosols do not act as potential IN in this case. Kuebbeler et al. (2012) and, indirectly, Cirisan et al. (2013) have demonstrated that only the indirect dynamical perturbation induced by SG may be capable of significantly perturb the number density of upper tropospheric ice particles, with decreased vertical velocities due to the enhanced atmospheric stabilization. As noted in Kuebbeler et al. (2012), the idea proposed in some studies that volcanic eruptions may enter larger and more abundant soluble aerosols into the upper troposphere (thus leading to enhanced ice crystals number concentrations) was indeed confirmed by ISCCP lidar measurements (Sassen et al. (2008)), whereas modeling studies found only a weak aerosol effect even in case of large aerosol perturbations (Karcher and Lohmann (2002); Lohmann and Feichter (2005)). However, it should be noted that in the case of explosive volcanic eruptions (contrary to SG) there are also solid ash particles injected in the lower stratosphere that will settle down below the tropopause (although with a rather short lifetime for the mass-dominant coarse mode), thus potentially contributing to some increase of the upper tropospheric IN population actually available for heterogeneous freezing. Gettelman et al. (2010) have shown that mineral dust particles can play an important role in cirrus cloud formation, because their ice active fraction may be rather large (>10% for a supersaturation ratio close to the homogeneous freezing threshold). However, this is not the case for the proposed SG, where the homogeneous freezing mechanism actually dominates.

Recent studies by Storelvmo et al. (2013) and Storelvmo et al. (2014) have quantified the direct radiative effects produced by seeding upper tropospheric cirrus ice clouds with large IN. Although this is not directly related to our specific discussion on SG side effects, it can be considered an indirect proof of the importance of correctly understanding the balance between the complex microphysical processes regulating the formation and growth of upper tropospheric ice particles.

We may conclude that the assumption of limiting our discussion to the indirect dynamical effect is a robust one and based on a sound physical basis. Kuebbeler et al. (2012) have calculated a longwave adjusted RF=-0.56 W/m^2 for all sky conditions under a SG injection of 5 Tg-SO_2/yr. However, we should keep in mind that some degree of uncertainty remains for the processes regulating the potential direct perturbation of upper tropospheric ice crystals through changes in the size distribution of sulfate aerosols acting as IN. As shown in Pitari et al. (2016b) for the atmospheric stabilization resulting from tropospheric aerosols by non-explosive volcanoes, the combined effect of the aerosol induced tropospheric decrease in temperature and updraft velocities produces a net global reduction of ice optical thickness in the upper troposphere of 1.0×10^{-3} at λ=0.55 μm, which then causes a radiative forcing of -0.08 W/m^2. This corresponds to an aerosol optical depth increase of 5.3×10^{-3} and an average surface cooling of 0.07 K. The same ULAQ-CCM module for ice crystals formation via homogeneous freezing has been applied to the SG case with stratospheric injection of 5 and 2.5 Tg-SO_2/yr, obtaining a globally averaged net radiative forcing of -0.47 and -0.35 W/m^2, respectively, in all sky conditions. These results are consistent with the above discussed findings of Kuebbeler et al. (2012).

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2.2.4 Methane

Another indirect effect of SG is a lifetime modification for many long-lived species. Among these species CH_4 is particularly important, due to its sensitivity on OH abundance and its impact on tropospheric chemistry. A CH_4 lifetime increase takes place for three main reasons (Pitari et al. (2014a)), all connected with a decrease in OH concentration, which represents the main sink for methane: (a) the surface cooling directly lessens the amount of water vapor in the troposphere, which in turns diminish OH concentration. (b) A decrease in tropospheric UV occurs in the tropics because of the stratospheric aerosols. This reduces the production of $O(^1D)$, which in turns decreases the amount of OH produced by the reaction $O(^1D) + H_2O$. (c) The increase of aerosol surface area density (SAD) enhances heterogeneous chemistry in the mid-upper troposphere, reducing the amount of NOx and O_3 production and thus of OH. The increased aerosol SAD produces a significant ozone depletion in the stratosphere whose effect is an increase of UV radiation able to reach the surface: however, such effect is overbalanced by the direct scattering of solar radiation, thus in the end the total amount of tropospheric UV is reduced (except over the polar latitudes)(Pitari et al. (2014a)).

In addition, it should be noted that the stratospheric aerosol heating rates produce a strengthening of the BDC, where more stratospheric air is transported from the stratosphere to the upper troposphere extra-tropics. Since the concentration of methane in the stratosphere is lower than in the troposphere, this strengthening of the BDC leads to a CH_4 decrease in the upper troposphere. All these effects together produce a longer lifetime of CH_4 that is estimated by the ULAQ-CCM to increase from 8 years for RCP4.5 to 9 years for G4 with injection of 5 Tg-SO_2/yr. According to the model, such a lifetime increase is estimated to produce a positive radiative forcing of +0.1 W/m^2 (Pitari et al. (2014a)).

2.3 To what extent may SG balance WMGHG RF?

Here we discuss how the estimated net RF from direct and indirect effects of SG should be compared with the positive RF associated with increasing WMGHG. The current IPCC scenarios for the next century will produce by 2100 a RF with respect to today?s levels of less than 1 W/m^2 (RCP2.6), $\sim$2.5 W/m^2 (RCP4.5), $\sim$4 W/m^2 (RCP6.0), and more than 6.5 W/m^2 (RCP8.5) (Moss et al. (2010); Meinshausen et al. (2011)). In the subsequent discussion, we choose not to consider the most optimistic, but probably not realistic, scenario RCP2.6 with a sharp RF reduction already before 2100.

The G4 experiment (Kravitz et al. (2011)) proposes a fixed amount of SO_2 to be injected in the stratosphere for the 2020-2070 period, in order to offset the positive RF by WMGHG. Therefore, a proper estimate for the magnitude of the required negative '(quasi) time-invariant' RF would be a number close to the average positive RF relative to 2020, during the whole period of the SG experiment (i.e., 2020-2070), although this implies an over-compensation of the positive RF from WMGHGs in the first two decades and an under-compensation afterwards.

A total estimate of the net RF from SG must take into account the wide range of factors discussed in the previous subsections. Here we would like to highlight that the relationship between the SO_2 amount and the subsequent AOD is non-linear, as larger amounts of SO_2 will produce larger aerosol particles and the aerosol scattering efficiency decreases. Furthermore, the gravita-

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tional settling becomes faster with increasing particle size, therefore reducing the stratospheric aerosol lifetime.

As highlighted in sub-section 2.1, another factor that may change the aerosol lifetime is the prolonged QBO westerly phase caused by SG, as discussed in Aquila et al. (2014). As showed in Pitari et al. (2016a) for explosive volcanic eruptions, a QBO with dominant easterly shear leads to a longer lifetime for the volcanic aerosol, due to a greater isolation of the tropical pipe. This helps confining the aerosols in an area where downward transport is not present. In a similar way, the extension of the lower stratospheric QBO westerly phase simulated by Aquila et al. (2014) leads to a longer aerosol lifetime. This result, however, could be partly canceled or even overcompensated if the microphysical effects of the QBO-dependent sulfur confinement in the tropical pipe were taken into account. Niemeier and Timmreck (2015) found that a locked QBO westerly phase globally produces a net decrease of the SG aerosol lifetime, because the tropical isolation leads to larger particles and subsequently to a more efficient gravitational settling.

Table 1 summarizes the RF values associated with SG found in several published studies. Aside from the direct effect of sulfate aerosol scattering, only the changes in cirrus ice particle formation and size may produce a RF of comparable magnitude. The indirect effects related to SG-induced changes in GHG concentrations (CH_4, O_3, stratospheric H_2O) are at least one order of magnitude smaller, so that we may assume that they are globally negligible with respect to the direct effect of SG aerosols and their indirect impact on ice cloudiness. Considering the results in Table 1, we found that the sum of all possible effects of both 2.5 and 5 Tg-SO_2/yr injection results in an overcompensation of the average positive RF over the 2020-2070 period, for the RCP6.0 and RCP4.5 cases. A still incomplete average compensation is achieved in the most pessimistic RCP8.5 case, more evident for the 2.5 Tg-SO_2/yr (+0.55 W/m^2).

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Table 1. A summary of the radiative forcing terms (W/m^2) of all mentioned SG effects, along with RCP4.5, RCP6.0 and RCP8.5 baseline scenarios: RF values are calculated as an average from 2020 to 2070. Data are taken or indirectly derived from Moss et al. (2010) (a), Pitari et al. (2014a) (b), Aquila et al. (2014) (c), Heckendorn et al. (2009) (d), Niemeier and Timmreck (2015) (e), Kuebbeler et al. (2012) (f), Pitari et al. (2014b) (g). The calculated average stratospheric AOD changes (at λ=0.55 μm) are 0.066±0.026 and 0.045±0.005 for 5 and 2.5 Tg-SO_2/yr injection, respectively. The asterisks denote a first approximation RF estimate, using a linear scaling of the 5 Tg-SO_2/yr value to the 2.5 Tg-SO_2/yr case using the ratio of the stratospheric AODs, when the model calculated RF value is not available for the 2.5 Tg-SO_2/yr case. Mean values and related uncertainties for the SO_4 direct RF are calculated from an average of the available published results.

Radiative forcing effects	RF (W/m^2) (SG → 5 Tg-SO_2/yr)	RF (W/m^2) (SG → 2.5 Tg-SO_2/yr)
Baseline RCP4.5 (2020-2070 mean) [WMGHGs+O_3+tropospheric aerosol]	+0.8 (a)	
Baseline RCP4.5 (2020-2070 mean) [WMGHGs+O_3+tropospheric aerosol]	+1.3 (a)	
Baseline RCP4.5 (2020-2070 mean) [WMGHGs+O_3+tropospheric aerosol]	+2.0 (a)	
Sulfate aerosol scattering	-1.45 ± 0.65 (b,c,d,e)	-1.15 ±0.15 (c)
UT cirrus ice changes	-0.55 (f)	-0.37 (f)*
GHGs (CH_4, O_3, stratospheric H_2O)	+0.1 (b,g)	+0.07 (b,g)*
Net [SG effects + RCP4.5]	-1.1	-0.65
Net [SG effects + RCP6.0]	-0.6	-0.15
Net [SG effects + RCP8.5]	+0.1	+0.55

3 Conclusions

Our assessment of the published literature on SG concludes that a SG has the potential to offset the RF due to GHG, even considering unwanted side effects such as cirrus changes and SG induced increases in GHG concentrations. The rather large uncertainty in the direct sulfate forcing calculated from independent values available in the literature should not surprise, due to model differences in the treatment of aerosol microphysics, latitude and altitude of SO_2 injection, QBO effects, changes in large scale transport produced by the aerosol heating rates and surface cooling. The uncertainties still present could hopefully be reduced in future with multi-model results obtained from a wide array of global models in coordinated projects, such as GeoMIP, with strict specifications regarding the SO_2 injection and aerosol microphysics and transport.

The net RF is considered here as a global average, providing no indication of how the regional climate would be effected by SG and how this would impact the hydrological cycle. Attention should also be used in studying the eventual side-effects of the termination of SG, so as to be sure that a powering down of the experiment would not have any negative side effect. Anyway, when comparing the SG techniques to others, it still appears to be one of the most feasible, taking into account its relatively

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high level of effectiveness and affordability (Robock et al. (2009); McClellan et al. (2012)). However, higher estimates on the SG costs have also been reported in the recent literature (Moriyama et al. (2016)), raising doubts on its affordability.

The above discussion highlights that still much is left to understand about the various effects on the climate of such a global endeavor. In no way such studies have the goal of deciding whether such a task has to be carried out. That remains a prerogative of populations and decision-makers. What we can do is offer a deep insight on all possible consequences, if ever the need arises for any geoengineering method to be deployed.

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