

Response to Reviewer 1 on “Sulfate geoengineering: a review of the factors controlling the needed injection of sulfur dioxide”

Comments are repeated in black italics. Replies are indicated in blue. Figures 1, 2a, 2b, 3a, 3b and 4 have been attached.

This is a review paper on sulfate geo-engineering and the factors controlling “the needed” injection of sulfur dioxide. The authors reviewed the direct radiative effect of sulfur injection that may lead to troposphere cooling and stratospheric warming, and the indirect radiative effect that caused by induced changes in ozone, CH₄, stratospheric water vapor, and upper tropospheric cirrus clouds. They compared the effect of GHG warming and the resulted changes by the direct and indirect effects of sulfate geo-engineering in order to estimate the best amount of sulfate to be injected. A critical review article that integrates and evaluates published literature is potentially very useful both for geo-engineering researchers and the broad atmospheric modeling community. Therefore, the effort the authors have made in this regard is greatly appreciated. However, I think the current manuscript needs to be substantially improved.

We thank the Reviewer for his encouraging general comment. As discussed below point-by-point, we have tried to incorporate all the Reviewer’s suggestions for improving the manuscript.

1) A few sections (2.1, 2.2.1-2.2.3) in the current review only passively summarize the findings from previous studies, but they don’t point out the weakness/gaps and suggest potential improvements and future directions. For example, many studies cited in the manuscript are based on the atmosphere-only model simulations forced by prescribed SST, so the interaction with the ocean is not considered. Another example is that the estimates from Cirisan et al. (2013) are based on box model simulations and radiative transfer model calculations, and it doesn’t consider the dynamical impact and the feedback to microphysics. Some careful discussions are needed for such cases.

A caution statement has been included in section 2.1 specifying the limitations of many of the atmosphere-only model simulations. Suggestions for future directions are also included (in particular the full coupling of SG aerosol with climate, stratospheric heating rates, QBO and inclusion of explicit microphysics). A caution statement is also included in the discussion of Cirisan et al. (2013) estimates. Final recommendations are given in the conclusions.

2) The authors did a good job in making connections between relevant studies, but in my opinion some of the discussions were presented with a bit too much detailed information (e.g. page 4 section 2.1), and the big picture was hidden behind some mixed topics. For example, I would suggest the authors to divide section 2.1 (and possibly 2.2.x) into two parts: 1) direct effects of sulfur injection (changes in microphysical properties, aerosol lifecycle, and optical properties) and the associated heating and cooling; 2) changes in circulation and its feedback. Also, as a review article, I think it is necessary to draw some schematic plots showing the major findings (mechanisms) from the literature (e.g. one each for sections 2.1, 2.2.1-2.2.4), so that the readers can have a quick overview of those studies. This is particularly important when the authors want to deliver comprehensive messages and opposing points from different studies.

We have followed the reviewer suggestion by splitting up section 2.1, with an introductory part on the direct effects of sulfur injection and a subsection 2.1.1 on the changes in circulation and its feedback. We have also introduced schematic summary plots for three sections: Fig. 1 and Fig. 2a-b in section

2.1, Fig. 3a-b for section 2.2.3, Fig. 4 for section 2.3. Sub-section 2.2.3 has also been split in three parts (2.2.3.1, 2.2.3.2, 2.2.3.3) discussing separately the processes of ice formation via homogeneous and heterogeneous freezing and finally the estimates of RF due to cirrus ice thinning. The figures are attached to this response.

3) *I think there are major flaws in Table 1 and the associated discussions (section 2.3). It seems to me that the authors are trying to project a net SG effect to compensate the RCP "forcing" (I think the authors should define their definition of forcing at the beginning) estimate. First, I am not clear how the authors derived the RCP RF numbers (not explicitly available in Moss et al. 2010), but it seems to me the "forcing" data presented in the paper are not calculated by CMIP models, but rather calculated using Integrated Assessment Models (IAM). Therefore, they might be very different from the real "forcing" estimated by the global climate models used in GeoMIP. Second, I think it's unacceptable to simply calculate the arithmetic mean the "forcing" numbers obtained from studies on different (direct/indirect) SG effects and the RCP estimates. Even if these numbers are estimated from the same model, the non-linear effect between the GHG warming, sulfate scattering, and cirrus cloud formation would result a very different estimate. I suggest to eliminate this part.*

Section 2.3 has been reorganized and changed following the reviewer criticism. Table 1 and its discussion has been eliminated. We now present a summary of the RF values associated to SG that were previously discussed in sections 2.1 and 2.2, using values published in the literature.

4) *Some additional literature need to be cited. For example, when discussing the impact on ozone, Tabazadeh et al. (2002) and Tilmes et al. (2008) should be cited and discussed.*

References:

Tabazadeh, A., Drdla, K., Schoeberl, M.R., Hamill, P. and Toon, O.B., 2002. Arctic "ozone hole" in a cold volcanic stratosphere. Proceedings of the National Academy of Sciences, 99(5), pp.2609-2612.

Tilmes, S., Müller, R. and Salawitch, R., 2008. The sensitivity of polar ozone depletion to proposed geoengineering schemes. Science, 320(5880), pp.1201-1204.

The ozone impact section has been completed with additional citations of published articles, including the ones suggested by the reviewer.

Minor issues: I saw quite some formatting problems and typos (especially RCP numbers in table 1). Please correct them.

Table 1 has been eliminated (see comment above).

Response to Reviewer 2 on “Sulfate geoengineering: a review of the factors controlling the needed injection of sulfur dioxide”

Comments are repeated in black italics. Replies are indicated in blue. Figure 4 is attached to the response to reviewer 1.

The paper summarizes geoengineering studies that discussed stratospheric SO₂ injections into climate models. The paper focusses only on a few studies. There are not that many studies in recent years that actually injected a fixed amount of SO₂ into the stratosphere. However, various studies used prescribed aerosol distributions. Those also contribute to the question of needed injections of sulfur dioxide. Therefore, I would recommend to extend this study to more papers, as listed below to justify the word “review” in the title. Also, I do not understand the last section of the paper and numbers in Table 1, and I think it needs more explanation.

We thank the Reviewer for his encouraging general comment. As discussed below point-by-point, we have tried to incorporate all the Reviewer’s suggestions for improving the manuscript.

Abstract: I disagree that the described technique would be planned for a timeframe of a few decades, while implementation of global measures of GHG emissions is achieved. This technique would likely have to be applied during and after global measures are implemented, and for a much longer period of time if aiming for temperature stabilization, since temperatures will still continue to rise after mitigation efforts have started. See for example Sanderson et al., 2016 (doi: 10.1002/2016GL069563), Tilmes et al., 2016 (doi:10.1002/2016GL070122); depending on the mitigation efforts, solar geoengineering may be required for a very long period of time.

Both abstract and introduction have been modified according to this comment. The introduction now states: **“Such geoengineering methods would need to be applied during and after global intergovernmental 10 measures on GHG emissions are implemented, in order to achieve surface temperature stabilization (Sanderson et al. (2016); Tilmes et al. (2016)).”**

Line 10: It will be very difficult to fine-tune amounts of sulfur dioxide emissions based on models, due to the range of climate sensitivity and differences in the response of surface temperatures to volcanic aerosols. All the different studies can do, is outline important factors that control the amount of sulfur dioxide to be injected.

Text modified according to this comment.

Page 2, Line 8. As commented above, it is misleading to assume that this technique would only be used between 2020 and 2070.

Text modified as suggested above.

Page 2, Line 21. Why would you only focus on the G4 type studies, why not extend this? Besides, there are other earlier studies that used fixed amounts of SO₂ injections, Rasch et al., 2006, and studies that prescribed sulfate aerosols based on fixed amounts of SO₂ injections, including Rasch et al, 2008 (doi:10.1029/2007GL032179), Tilmes et al., 2009 (doi:10.1029/2008JD011420), Tilmes et al., 2012 (doi:10.5194/acp-12-10945-2012). Those and others may be included in the review.

The reviewer suggestion has been followed in the revised version, including earlier studies with fixed amounts of SO₂ injections and also including a documented G3-type study in the ozone section.

Line 26: You can also add Niemeier et al., 2011 (doi:10.1002/asl.304), and Niemeier et al., 2013 (doi:10.1002/2013JD020445).

References added.

Page 3: Direct forcing of stratospheric sulfate: References in the first paragraph are very old and by now there are more recent papers describing that the cooling effect after Mt. Pinatubo was actually much smaller (at most 0.3 C), IPCC 2015, Canty et al., 2013 (doi:10.5194/acp-13-3997-2013). Also the radiative forcing seems to be largely overestimated in the study by Minnis et al., 1993.

References updated for the globally averaged temperature change after Pinatubo. The text now states: **“This was calculated as a monthly mean for September 1992, compared to pre-Pinatubo levels. However, more recent results with detrended analyses (Canty et al. (2013)) have shown that the Pinatubo volcanic impact on surface temperatures was probably overestimated by about a factor of 2, with a cooling estimate of 0.14 K and 0.32 K, globally and over land, respectively.”** The estimate of Stowe et al. (1992) (~2.5 Wm⁻²) is used for the net TOARF.

Page 3, Line 28: The range in radiative response was likely due to the differences in AOD of the models. However, even with the same AOD distribution, models may have very different radiative responses, see for example Neely et al., 2015 (doi:10.5194/gmdd-8-10711-2015), just comparing 2 CESM versions with different radiation schemes.

Text modified accordingly. We added the lines: **“The different results are mainly dependent on the (calculated, or imposed in one case) different aerosol optical depth (AOD) and size distribution among models. It should also be considered that, in general, even with the same AOD distribution, models may produce different radiative responses depending on the adopted radiation scheme (Neely et al. (2016)).”**

Page 3, Line 13: please change to “a series of factors”.

Changed.

Section 2.2.1 Ozone. This section only summarizes findings from one paper, this is not a review. Heckendorn et al., 2009 (doi:10.1088/1748-9326/4/4/045108) and Tilmes et al., 2009 (doi:10.1029/2008JD011420), have discussed changes in ozone due to solar geoengineering.

In the original manuscript we were focusing only on the topic of the indirect RF due to ozone changes, which was extensively reported only in Pitari et al. (2014). But we agree that in a review article the discussion should be extended to all relevant physical and chemical processes involved. A more complete coverage of the recent literature for the SG effects on stratospheric ozone is now made in the revised manuscript. We have added the following phrases: **“Early studies of the potential impact of SG on stratospheric ozone are those of Tilmes et al. (2008), Tilmes et al. (2009) and Heckendorn et al. (2009). Tilmes et al. (2008) focus on polar ozone and estimate that SG could favor stratospheric ozone destruction and delay the recovery of the Antarctic ozone hole by 30-70 years. In addition, this ozone depletion produces a significant increase of erythemal surface UV,**

up to 5% in mid- and high latitudes and 10% over Antarctica (Tilmes et al. (2012)). The polar ozone depletion is favored by enhanced NO_x removal via heterogeneous chemical reactions on the surface of stratospheric sulfate aerosols, as in the case of major volcanic eruptions taking place with high atmospheric levels of chlorine and bromine species (Tabazadeh et al. (2002)). Tilmes et al. (2009) and Heckendorn et al. (2009) analyze the SG impact in chemical ozone loss rates and find that the chemical ozone changes are significantly impacted by the strong reduction of the NO_x cycle, due to the efficient NO_x to HNO₃ conversion on the surface of sulfate aerosols. The NO_x depletion, in turn, favors an increase of HO_x, Cl_x and Br_x loss rates: the net effect on column ozone column will then be time-dependent and regulated by the amount of halogen species in the lower stratosphere. Heckendorn et al. (2009) have calculated a global ozone reduction of 4.5% (i.e., ~13 DU), for an injection of 10 Tg-SO₂/yr and assuming halogen concentrations appropriate for year 2000. Pitari et al. (2014) have run the GeoMIP G4 experiment from 2020 to 2070: despite the constant stratospheric aerosol loading, the magnitude of the geoengineering aerosol induced ozone depletion is found to decrease in time, due to the decreasing atmospheric concentration of chlorine and bromine species. Two of the models used in this study (ULAQ-CCM and MIROC-ESM-CHEM) even show a global ozone increase starting from about 2050, when the NO_x driven chemical ozone increase is no longer overbalanced by the HO_x, Cl_x and Br_x driven ozone loss.”

Page 5, Line 13: Do the numbers -1.1 to -2.1 DU include the model that did not consider heterogeneous chemistry? How do those numbers compare to earlier studies? Same for the RF, what models are included in this number?

A more complete and precise discussion is now made in the revised manuscript, with appropriate citations to previous studies.

Section 2.2.3. Do you mean “Upper tropospheric ice”?

Thank you for catching this typo. Corrected.

Page 8, Line 12; Please note, tropospheric UV shows a net reduction in the tropics, correctly stated in the text. However, this is not the case of mid- and high latitudes. Methane lifetime is mostly influenced from OH changes in the tropics, therefore the methane lifetime is increased with geoengineering.

A sentence has been added to make it clear that the high-latitude UV increase has little effect on the methane lifetime.

*Line 23: typo: today’s, also what do you mean by today’s levels, what period? Could you explain the numbers given in Section 2.3 and Table 1? For example, to offset certain levels of RF, one would need to identify how much sulfur injection is required, which is model depended. For instance, Niemeier and Timmreck, 2015, calculated an efficiency of 0.30 – 0.35 W/m² per TgS injection. Since 5 Tg SO₂ are equal to 2.5 TgS, this results in about 0.3*2.5 = 0.75 W/m² per 5 Tg SO₂ injection. Can you do the same calculations for the other studies? It is not clear how you get to the value of -1.45 W/m² +/- 0.65 in this study. Also, for example the RF of RCP 4.5 between 2020 and 2070 is about 2.2-2.3 W/m². Where does the number in Table 1 (0.8 W/m²) come from? If the RF needs to be set off by geoengineering in 2070, much more forcing is required than 0.8 W/m².*

Today's level is now specified: RF is estimated for year 2100 relative to year 2011. Table 1 has been eliminated. We agree that our attempt to quantify a net residual from the RCP net RFs over the "50 year period of SG application" minus the net RF from SG is not clear and not fully justified, on light of the previous criticisms. For this reason, we simply summarize the IPCC findings on the net RFs following different RCPs and we present our findings on the breakdown per component of the SG RF in a "stand-alone" figure, taking into account the estimates published in the recent literature and separately discussed in sections 2.1 and 2.2.

For the cirrus forcing, why do you only state one number for cirrus impacts and not the lower number from Pitari et al., 2016b? Particle sizes from sulfate geoengineering are likely not large enough to have any significant effect, while dust particles have a larger effect. In Table 1, at least give a range for cirrus cloud effects.

The RF summary plot (Fig. 4) now includes whiskers for all the components, including cirrus ice. We thank the reviewer for the specific suggestion. By the way, SG particles are inefficient IN, mainly because they are supercooled liquid particles, contrary to (solid) dust particles.

Response to Reviewer 3 on “Sulfate geoengineering: a review of the factors controlling the needed injection of sulfur dioxide”

Comments are repeated in black italics. Replies are indicated in blue.

This paper presents a review of studies of sulfate geoengineering (SG). The paper selects results from a wide body of literature, of which some significant works have been left out. I agree with referee comment 1 (RC1) that more information should be given about the limitations of the studies presented, and the relative strength of conclusions possible. I have attached an annotated PDF with corrections and comments, which I summarize and expand on here.

We thank the Reviewer for his constructive comments. As discussed below point-by-point, we have tried to incorporate all the Reviewer’s suggestions for improving the manuscript.

Page 2, lines 7-8: This is a dangerously false statement. If SG were applied only during the transition period to clean energy source, its abrupt halt would trigger catastrophically rapid global warming, since the negative forcing of stratospheric sulfates would be removed within a few years, while the positive forcing of carbon dioxide would remain for thousands of years. Unless humans can remove much of the carbon dioxide from the atmosphere that we have added over 170 years thus far, SG would have to be applied indefinitely on human timescales. As the paper alludes to, carbon air capture remains a very elusive and energy intensive process, and it is far from clear that it would be viable on a large scale by 2070.

Following the same recommendation of the second reviewer, we have cut this statement from both introduction and abstract.

Page 2, line 23: What is meant by “the GeoMIP experiment Robock et al. (2011)”, in contrast to “the GeoMIP experiment G4” on line 21?

That was a typo: “G3” is missing. Corrected, with additional references.

Page 3, line 6: It should be clarified that the 0.5 C drop in global average temperature was a monthly average, not an annual average.

Following the same criticism by the second reviewer, we have corrected this statement, with additional references.

Page 4, lines 7 and 12: clarify at what latitude(s) SO₂ was injected, and how emissions were zonally distributed.

Clarified, for both Aquila et al. (2014) and Niemeier and Timmreck (2015).

Page 4, line 8: “proportionally” implies a linear relationship of aerosol mass injected to the period of the westerly phase. This does not see right if a permanent westerly is achieved with a finite injection rate.

We agree that “proportional” is not the right word to describe this effect. Corrected as follow: **“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. (2014a).”

Page 5, section 2.2.1: It is unclear that the attribution of reduction in O₂ photolysis as the “main” cause of the reduction in column ozone is reasonable absent experiments in which O₂ photolysis rates are unchanged by sulfate AOD. The catalytic loss rates are proportional to the amount of ozone present, so might be larger if ozone production were not reduced. The later discussion that column ozone increases with SG after 2060, when chlorine and bromine are reduced, makes this point less convincing.

We admit there was some confusing statements in the original manuscript. We have simplified our sentence as follows: **“The models used in the G4 experiment showed significant changes in the ozone profile, with a decrease in the tropical column between 100 and 30 hPa in the tropics, for the combined effects of enhanced upwelling and losses in the chemical cycles.”**

I agree with RC2 that Table 1 is unclear and requires substantial further explanation.

Table 1 has been eliminated. We agree that our attempt to quantify a net residual from the RCP net RFs over the “50 year period of SG application” minus the net RF from SG is not clear and not fully justified, on light of the previous criticisms. For this reason we simply summarize the IPCC findings on the net RFs following different RCPs and we present our findings on the breakdown per component of the SG RF in a “stand-alone” figure, taking into account the estimates published in the recent literature and separately discussed in sections 2.1 and 2.2.

I have included a few typographical corrections as well in the annotated PDF.

The sticky notes on the original pdf document have been properly considered in the revised manuscript.

Finally, there are a number of additional studies that could be discussed in this review. RC1 and RC2 have identified a number of these. I would suggest at least including some discussion of these papers:

Tilmes, S., R. Müller, and R. Salawitch (2008), The sensitivity of polar ozone depletion to proposed geoengineering schemes, Science, 320(5880), 1201–1204, doi:10.1126/science.1153966.

Tilmes, S. et al. (2013), The hydrological impact of geoengineering in the Geoengineering Model Intercomparison Project (GeoMIP), J. Geophys. Res-Atmos, 118(1), 11036–11058, doi:10.1002/jgrd.50868.

Tilmes, S., B. M. Sanderson, and B. C. O’Neill (2016), Climate impacts of geoengineering in a delayed mitigation scenario, Geophys. Res. Lett., 43(15), 8222–8229, doi:10.1002/2016GL070122.

These (and other references to relevant SG studies) are included in the revised manuscript.

Please also note the supplement to this comment: <http://www.atmos-chem-phys-discuss.net/acp-2016-985/acp-2016-985-RC3-supplement.pdf>

The sticky notes on the original pdf document have been properly considered in the revised manuscript.

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

Daniele Visioni^{1,2}, Giovanni Pitari¹, and Valentina Aquila^{3,4}

¹Department of Physical and Chemical Sciences, Università dell'Aquila, 67100 L'Aquila, Italy

²CETEMPS, Università dell'Aquila, 67100 L'Aquila, Italy

³GESTAR/Johns Hopkins University, Department of Earth and Planetary Science, 3400 N Charles Street, Baltimore, MD 21218, USA

⁴NASA Goddard Space Flight Center, Code 614, Greenbelt, MD 20771, USA

Correspondence to: Daniele Visioni (daniele.visioni@aquila.infn.it)

Abstract.

Sulfate geoengineering has been proposed as an affordable and climate-effective ~~means-for-mean to~~ 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-WMGHG_s (CO₂ in particular) technique would likely have to be applied during and after global intergovernmental measures on emissions of WMGHGs are implemented, in order to achieve surface temperature stabilization. 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 longwave 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 CH₄ in connection with OH changes through several photochemical mechanisms. ~~A direct comparison of the net effects of WMGHG increase with direct~~ Direct and indirect effects of sulfate geoengineering ~~may help fine-tune the best amount of sulfate~~ both concur to determine the atmospheric response. A review of previous studies on these effects is presented here, with an outline of the important factors that control the amount of sulfur dioxide to be injected in an eventual realization of the experiment. However, we need to take into account ~~large uncertainties that atmospheric models used for these studies have shown a wide range of climate sensitivity and differences in the response to stratospheric volcanic aerosols. In addition, large uncertainties exist~~ 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~~ surface warming 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 (IPCC (2007)). Such a need is made even more pressing if we look at the projections for the next century. The ~~InterGovernmental~~ Intergovernmental Panel on Climate Change (IPCC) has built various Representative Concentration Path-

ways (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 5 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, even with the most optimistic emission scenario, a sudden reversing of the temperature trend is not expected (IPCC (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. 10 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))~~ during and after global intergovernmental measures on GHG emissions are implemented, in order to achieve surface temperature stabilization (Sanderson et al. (2016); Tilmes et al. (2016)). 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 CO₂ 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 (SRM) techniques, whose aim is to decrease the amount of incoming 15 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 (SO₂) 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. (2011); Kravitz et al. (2012); Kravitz et al. (2013)) 25 ~~-chemistry-climate models and atmosphere-ocean coupled models have been used to explore the radiative, chemical and dynamical modification of climate by SO₂ injection. Several studies were conducted to compare a control simulation ensemble under the IPCC scenario RCP4.5 (Taylor et al., 2012) 2(Taylor et al. (2012)) and a sulfate geoengineering simulation. In this review we summarize the direct and indirect climate effects of a constant stratospheric injection of SO₂, such as the one prescribed by the GeoMIP experiment G4 ~~, where 5 Tg/year of SO₂ were injected in the tropical lower stratosphere from 2020 to 2070 (Pitari et al. (2014) Aquila et al. (2014a)), rather than (Pitari et al. (2014); Aquila et al. (2014a)) and in earlier studies (Rasch et al. (2008); Tilmes et al. (2009); Tilmes et al. (2012)) or of a time-varying SO₂ injection, such as in the GeoMIP experiment Roboek et al. (2011), where G3 (Pitari et al. (2014)). In this case~~ the amount of the injected SO₂ changes year-by-year in order to keep the top-of-atmosphere (TOA) radiative balance constant ~~. This is because the G4 (Roboek et al. (2011); Kravitz et al. (2011)). The G4-type~~ approach (even if with different amounts of constant SO₂ injection) has been used and 30 documented in a wider number of studies (see also Heckendorn et al. (2009); ~~English et al. (2012); Niemeier et al. (2011);~~~~

[English et al. \(2012\)](#); [Niemeier et al. \(2013\)](#); Niemeier and Timmreck (2015)).

The direct effect of an injection of SO₂ is an increase in the local concentration of optically active H₂O-H₂SO₄ 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
5 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)).

2 Review of radiative forcing effects

10 2.1 Direct forcing of stratospheric sulfate

The underlying physical processes behind the injection of SO₂ into the atmosphere have been widely studied thanks to the various explosive volcanic ~~eruption~~ ~~eruptions~~ of the 20th century. For instance, after the Mount Pinatubo eruption of June 1991, ~~where~~ ~~when~~ 7 to 10 Tg-S ~~where~~ ~~were~~ injected into the stratosphere (Read et al. (1993); Krueger et al. (1995)), a ~~significant drop in surface temperature of about 0.5 K~~ ~~sharp reduction in the TOA net radiative flux~~ was observed in the year following
15 the eruption (~~Dutton and Christy (1992)~~), ~~along with a sharp reduction in the TOA net radiative flux (3 to 10~~ ~~~2.5~~ W/m²) ~~right after the eruption (?)~~. (Stowe et al. (1992)), as well as a significant drop in global surface temperatures of about 0.5 K (Dutton and Christy (1992)). This was calculated as a monthly mean for September 1992, compared to pre-Pinatubo levels. However, more recent results with detrended analyses (Canty et al. (2013)) have shown that the Pinatubo volcanic impact on surface temperatures was probably overestimated by about a factor of 2, with a cooling estimate of 0.14 K and 0.32 K, globally
20 and over land, respectively.

These effects can be explained by SO₂ oxidation into SO₄ followed by the formation of H₂O-H₂SO₄ 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
25 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~~ ~~factors~~ must be taken into account, complicating the analogy between ~~these~~ ~~this~~ 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)
30 analyzed a series of ~~different~~ geoengineering experiments accounting for the different factors previously mentioned. Their results show that the relation between injected SO₂ and the ~~resulting~~ 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 SO₂ lead to the formation of larger aerosol particles by gas condensation, which are rapidly removed from the stratosphere by gravitational settling ~~(see~~

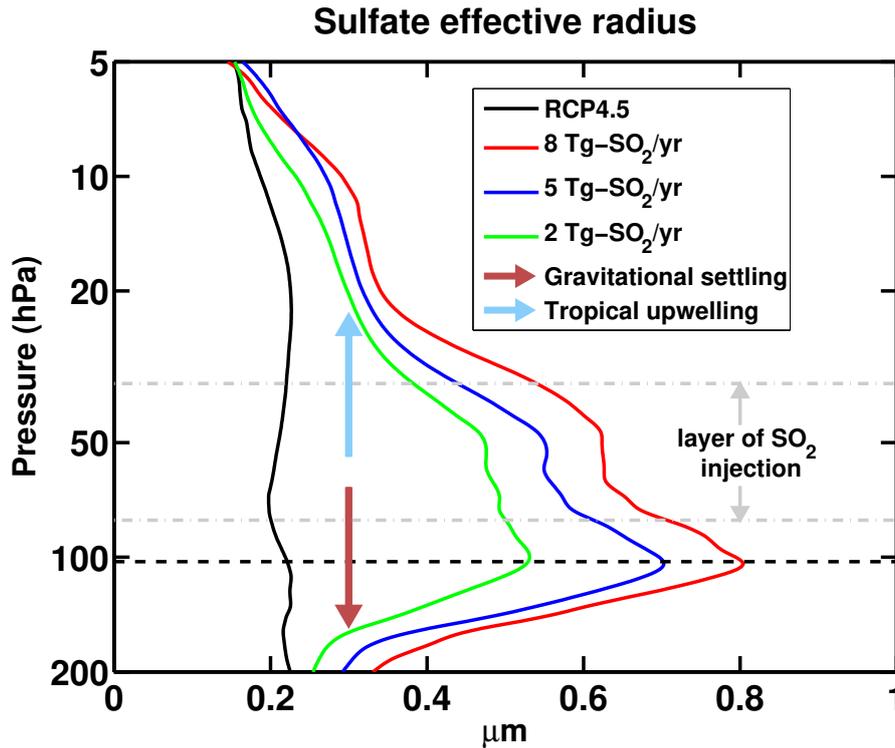


Figure 1. Annual averaged vertical profiles of aerosol effective radius (μm) in the tropical stratosphere (25S-25N), with increasing geoengineering injection of SO_2 (see legend). The heavy dashed line indicates the mean tropical tropopause. Profiles are calculated in the University of L'Aquila Chemistry-Climate Model (ULAQ-CCM), which includes explicit gas-particle conversion and aerosol microphysics (Pitari et al. (2014)).

Fig. 1, with calculated vertical profiles of the aerosol effective radius).

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 (English et al. (2012)). The highest burden to injection ratio is achieved for stratospheric injections between 30N and 30S (English et al. (2012)), because gas condensation and particle coagulation are both reduced with SO_2 injection spanning over a broader latitude. The altitude also plays a significant role in determining the aerosol lifetime, due to a faster sedimentation removal in the upper troposphere (UT) when the sulfur injection is localized closer to the tropical tropopause layer (TTL) (Aquila et al. (2014a)).

As shown in Pitari et al. (2014), the injection of 5 Tg- SO_2 /yr produces, according to the models used in the experiment G4, a net TOA radiative forcing (RF) between of -1.54 W/m^2 , -1.27 W/m^2 , -1.31 W/m^2 and -0.73 W/m^2 , for ULAQ-CCM, GEOSCCM, GISS-E2-R and MIROC-ESM-CHEM, respectively (Pitari et al. (2014) for model description and details). 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. (2014)~~ It should also be considered that, in general, even with the same AOD distribution, models may produce different radiative responses depending on the adopted radiation scheme (Neely et al. (2016)). Other RF values are available from literature, for a variety of conditions of sulfur injection (amount and altitude, mainly). With a linear scaling to 5 Tg-SO₂/yr (in case of different injection values), we get the following values: -1.13 W/m² (Heckendorn et al. (2009)); -1.17 W/m² (Niemeier et al. (2011)); -1.53 W/m² (Kuebbeler et al. (2012)); -1.4 W/m² and -1.0 W/m² (Aquila et al. (2014a)); -0.87 W/m² (Niemeier and Timmreck (2015)). In two cases, the forcing value was reported as the surface shortwave (SW) RF (Heckendorn et al. (2009); Niemeier et al. (2011)); it has been converted to a net TOA RF by scaling the SW surface value with a factor (25-8)/20, where 25, 20 and 8 are the approximate factors to derive TOA SW, surface SW and TOA adjusted longwave (LW) RFs from the stratospheric AOD. From these RF values available in the literature, we may derive a mean value of -1.19 ± 0.27 W/m².

2.1.1 Changes in circulation and its feedback

While on the one hand these results show that SG leads to the desired effect of (at least partially) offsetting the positive RF of increasing well mixed greenhouse gases (WMGHG), on the other hand they show that SG ~~side effects such as effects, such as~~ the 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. (2016b)). The tropical lower stratospheric warming induces a significant 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. (2014)~~-BDC) (Pitari et al. (2014); Pitari et al. (2016a)).

~~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 quasi-biennial oscillation (QBO) under geoengineering conditions have been analyzed in the aforementioned study by Aquila et al. (2014a) using the NASA Goddard ~~climate-chemistry-coupled chemistry-climate~~ model (GEOSCCM), which includes an internally generated QBO. Four different experiments were designed, using 5 Tg-SO₂/yr for the first two and 2.5 Tg-SO₂/yr for the others, injected at different altitudes (16-25 km and 22-25 km; both at the equator and 0° longitude in a single lat/lon box). 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 with an increasing~~ stratospheric SO₄ mass burden (ranging from 1.5 Tg-S for the 16-25 km injection of 2.5 Tg-SO₂/yr to 4.7 Tg-S for the 22-25 km injection of 5 Tg-SO₂/yr).

Niemeier and Timmreck (2015) also mention a perturbation of the QBO in SG simulations performed with the ECHAM-HAM model. ~~Their simulation~~ This was an ensemble of simulations with variable SO₂ injection (1-100 Tg-S/yr), altitude and latitude of injection (60 hPa and 30 hPa; Eq-2.8°N; 5°S-5°N; 30°S-30°N; all in a single longitude box centered at 122.3°E). Their

simulations 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~~ 8 Tg-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. (2014a). The overall conclusion of both these studies is that a stratospheric sulfur injection could dramatically alter the

5 QBO periodicity, up to producing a permanent westerly phase in the lower stratosphere, thus reducing the meridional transport efficiency (Trepte and Hitchman (1992)).

The SO₄ stratospheric lifetime in the simulations included in Aquila et al. (2014a) 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-SO₂/yr case with respect to the 2.5 Tg-SO₂/yr injection case (~1.9 years versus

10 ~1.7 years with injection in the 22-25 km layer and ~1.25 years versus ~1.2 years with injection in the 16-25 km layer). The higher heating rates produced by the aerosol in the ~~5Tg-SO~~ 5 Tg-SO₂/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. (2016b)). This reduces the amount of aerosol that may be transported downwards across the extra-tropical tropopause in the lower branch of the BDC. A compact summary of

15 all these feedback mechanisms is presented in Fig. 2 (superimposed to the calculated sulfate mass density anomaly due to an injection of 5 Tg-SO₂/yr).

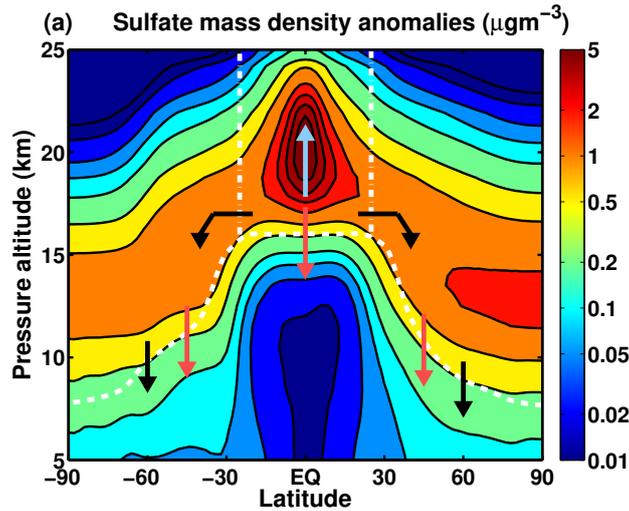
The prolonging of the aerosol lifetime found by Aquila et al. (2014a), however, could be canceled if the microphysical effects of the QBO-dependent sulfur confinement in the tropical pipe were taken into account. In the simulations by Niemeier and Timmreck (2015) using the ECHAM-HAM model, which includes a representation of aerosol microphysics, the enhanced

20 aerosol tropical confinement under ~~conditions~~ condition of a locked QBO westerly phase in the lower stratosphere decreases the SG aerosol lifetime, this is because the tighter tropical confinement of the aerosol also leads to larger particles and therefore a more efficient gravitational settling (U. Niemeier, personal communications) .(see Fig. 2b).

Many of the previous cited studies have focused on specific aspects of formation, transport and removal of stratospheric aerosols under geoengineering conditions. As noted above, significant feedback mechanisms exist among the magnitude and location

25 of SO₂ injection, aerosol microphysics, background stratospheric dynamics, aerosol induced surface cooling and stratospheric heating rates, as well as induced changes in the stratospheric circulation and strat/trop exchange. This means that a significant improvement on the knowledge of direct and indirect effects of SG may be obtained through model experiments designed in such a way that all these aspects are explicitly considered and interacting with each other. One important limitation of many of the above cited studies is the use of atmosphere-only models forced by prescribed sea surface temperatures (SST), so that an

30 explicit interaction of geoengineering aerosols with surface ocean is not considered. A missing explicit aerosol microphysics is another limitation for some of these studies: in this case, the increased gas-particle conversion cannot feedback on the aerosol size distribution shape and finally on the particle sedimentation rate and aerosol optical properties for the radiative transfer calculations.



(b) Summary of SO_2 injection feedback mechanisms

	Dynamical effect	With increasing SO_2 injection:	Sulfate lifetime & optical depth
↓	Gravitational settling	Increases [Enhanced gas-particle conversion: larger particles]	Decrease
↔	Isentropic poleward transport & strat-trop exchange	Decreases [Prolonged QBO W phase: higher tropical confinement]	Increase
↓	Tropical gravitational settling	Increases [Higher sulfur confinement due to QBO effect: larger particles]	Decrease
↑	Tropical upwelling	Increases [Enhanced aerosol heating rates]	Increase

Figure 2. Panel (a): annually and zonally averaged sulfate mass density calculated anomalies ($\mu\text{g/m}^3$), due to a geoengineering injection of 5 Tg- SO_2/yr , with respect to a RCP4.5 background atmosphere. The aerosol mass density distribution is calculated in the Goddard Earth Observing System Chemistry Climate Model (GEOSCCM), with SG treated as described in Pitari et al. (2014). Arrows superimposed to the aerosol distribution indicate the main transport pathways of the aerosol particles, as explained in panel (b). The white dashed line shows the mean tropopause; the dash-dotted white lines highlight the stratospheric tropical region. The sensitivity of each dynamical effect to the SO_2 injection is highlighted in panel (b), along with the physical mechanisms driving the perturbation and the net effect on sulfate lifetime and optical depth.

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~~ SG of counteracting the positive RF from WMGHGs.

2.2.1 Ozone

Early studies of the potential impact of SG on stratospheric ozone are those of Tilmes et al. (2008), Tilmes et al. (2009) and Heckendorn et al. (2009). Tilmes et al. (2008) focus on polar ozone and estimate that SG could favor stratospheric ozone destruction and delay the recovery of the Antarctic ozone hole by 30-70 years. In addition, this ozone depletion produces a significant increase of erythemal surface UV, up to 5% in mid- and high latitudes and 10% over Antarctica (Tilmes et al. (2012)). The polar ozone depletion is favored by enhanced NO_x removal via heterogeneous chemical reactions on the surface of stratospheric sulfate aerosols, as in the case of major volcanic eruptions taking place with high atmospheric levels of chlorine and bromine species (Tabazadeh et al. (2002)).

Tilmes et al. (2009) and Heckendorn et al. (2009) analyze the SG impact in chemical ozone loss rates and find that the chemical ozone changes are significantly impacted by the strong reduction of the NO_x cycle, due to the efficient NO_x to HNO_3 conversion on the surface of sulfate aerosols. The NO_x depletion, in turn, favors an increase of HO_x , Cl_x and Br_x loss rates; the net effect on the ozone column will then be time-dependent and regulated by the amount of halogen species in the lower stratosphere. Heckendorn et al. (2009) have calculated a global ozone reduction of 4.5% (i.e., ~ 13 DU), for an injection of 10 Tg- SO_2 /yr and assuming halogen concentrations appropriate for the year 2000. Pitari et al. (2014) have run the GeoMIP G4 experiment from 2020 to 2070: despite the constant stratospheric aerosol loading, the magnitude of the geoengineering aerosol induced ozone depletion is found to decrease in time, due to the decreasing atmospheric concentration of chlorine and bromine species. Two of the models used in this study (ULAQ-CCM and MIROC-ESM-CHEM) even show a global ozone increase starting from about 2050, when the NO_x driven chemical ozone increase is no longer over-balanced by the HO_x , Cl_x and Br_x driven ozone loss.

Model simulations in Pitari et al. (2014) 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 an average total change SG induced perturbation of -2.8 ± 3.0 DU is calculated in the global mean ozone column from -1.1 to -2.1 DU is calculated for the 2040-2049 decade, considering decadal averages from 2020 to 2070 for the 4 models that ran the G4 experiment (ULAQ-CCM, GEOSCCM, GISS-E2-R and MIROC-ESM-CHEM) and for the two models that ran the G3

experiment (ULAQ-CCM and GISS-E2-R). In terms of RF this produces a rather small negative result, ~~ranging from -0.028 to -0.036~~ of the order of -0.04 W/m^2 ~~∴ RF=-0.045±0.035 W/m², with the same decadal averages used for the global mean ozone column change.~~

- 5 ~~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 NO_x 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

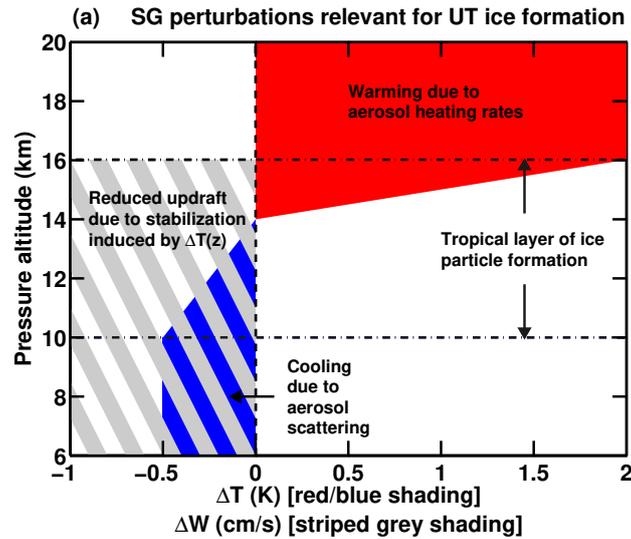
SG is expected to increase stratospheric water vapor ~~concentrations~~ concentration by warming the TTL ~~temperature~~. In the
10 stratosphere, the water vapor concentration is regulated by the TTL temperature (Dessler et al. (2013)), combined with methane oxidation. The ~~warmer~~ higher the TTL temperatures, 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
15 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
20 case. Pitari et al. (2014) 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 ~~0.02-0.08~~ to 0.35 ppmv. This in turn produces a net positive RF ~~ranging between +0.004 and +0.077~~ =0.055±0.025
25 W/m², considering decadal averages from 2020 to 2070 for the 3 of the 4 models that ran the G4 experiment (ULAQ-CCM, GEOSCCM, and MIROC-ESM-CHEM). The fourth model (GISS-E2-R), on the other hand, predicts a TTL cooling with a decreased amount of stratospheric H₂O and ~~then thus~~ 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 tropospheric ice

2.2.4 ~~Upper stratospheric ice~~

30 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 updraft. 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

5 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. Fig. 3 presents a compact summary of the dynamical perturbations induced by SG and relevant for the ice particle formation via homogeneous freezing. Lower vertical velocities force a decrease in ice crystals number concentration due to the decreasing water vapor transport from below, with consequent lower supersaturation. The temperature dependence is inverse, because lower temperatures allow for more ice crystals, due to the slower depositional growth and the higher nucleation rate (Kuebbeler et al. (2012)).

(b) Summary of SO₂ injection feedback mechanisms

Thermal-Dynamical effect	With increasing SO ₂ injection:	UT ice optical depth
Lower stratospheric & uppermost tropospheric warming	Increases [Enhanced aerosol heating rates due to LW radiation absorption]	Decreases [Faster depositional growth and lower nucleation rates]
Tropospheric cooling	Increases [Enhanced aerosol SW radiation scattering]	Increases [Slower depositional growth and higher nucleation rates]
Vertical velocity and water vapor updraft	Decreases [Enhanced tropospheric stabilization due to induced T(z) changes]	Decreases [Lower supersaturation: less ice crystals can nucleate]
Aerosol gravitational settling	Increases [Enhanced gas-particle conversion: larger particles]	Increases (?) [More UT sulfate aerosols, but inefficient IN for heterogeneous freezing]

Figure 3. Panel (a): schematic profile changes of upper troposphere-lower stratosphere temperature (K) and UT vertical velocity (cm/s) in the tropics, due to a geoengineering injection of 5 Tg-SO₂/yr. The perturbation scheme is based on the findings of Kuebbeler et al. (2012), Pitari et al. (2016c) and Pitari et al. (2014). The dash-dotted black lines indicate the region of ice particle formation (up to the mean tropopause). The sensitivity of each thermal-dynamical effect to the SO₂ injection is highlighted in panel (b), along with the physical mechanisms driving the perturbation and the net effect on UT ice optical depth.

2.2.3.1 Ice formation via homogeneous freezing

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 $T=220 \text{ K}$ 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 ~ 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 $0.5 \mu\text{m}$), 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 $+0.02 \text{ W/m}^2$ 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 that the effect of a perturbed aerosol size distribution on the ice particle population formed via homogeneous freezing is of secondary importance. It should be considered, however, that the estimates from Cirisan et al. (2013) are based on box model simulations and radiative transfer model calculations, and do not consider the dynamical impact and the feedback to microphysics.

2.2.3.2 Ice formation via heterogeneous freezing

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)~~ (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 lead~~ to enhanced ice ~~crystals number concentrations~~) crystal 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.

15

2.2.3.3 RF estimates from cirrus ice thinning

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 LW TOA RF=-0.56-0.51~~ W/m² for ~~all-sky conditions~~ cloud adjustment due to optically thinner cirrus, under a SG injection of 5 Tg-SO₂/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. In addition, as noted by the author themselves, one limitation of the study by Kuebbeler et al. (2012) is that sea surface temperatures were prescribed. The SG induced cooling of the surface would on one hand enhance the atmospheric stabilization and then further reduce the vertical updraft and cirrus ice optical depth (see Fig. 3), but on the other hand it would contribute to cool the whole troposphere, thus favoring additional ice crystals formation (see Fig. 3). Although Kuebbeler et al. (2012) suggest that in principle it would be important to redo the simulations with a mixed layer ocean, on the other hand they conclude that the overall difference in the GCM response would be small in term of UT ice anomalies.

As shown in Pitari et al. (2016c) 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 $\lambda=0.55 \mu\text{m}$, which then causes a radiative forcing of -0.08 W/m². 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-SO₂/yr, obtaining a globally averaged ~~net radiative forcing of -0.47 and -0.35~~ LW TOA RF=-0.45 W/m² , respectively, due to optically thinner cirrus, consistent with the findings of Kuebbeler et al. (2012). A corresponding net TOA RF=-0.30 W/m² was calculated in all sky conditions. ~~These results are consistent with the above~~

~~discussed findings of Kuebbeler et al. (2012)~~, with the SW RF=0.15 W/m² (i.e., 34% of the absolute LW RF). If this same SW/LW RF fraction is applied, a net TOA RF=-0.34 W/m² is obtained for Kuebbeler et al. (2012), for the cloud adjustment due to optically thinner cirrus.

5

2.2.4 Methane

Another indirect effect of SG is a lifetime modification for many long-lived species. Among these species CH₄ is particularly important, due to its sensitivity ~~on to~~ OH abundance and its impact on tropospheric chemistry. A CH₄ lifetime increase takes place for three main reasons (Pitari et al. (2014) Aquila et al. (2014b)), 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~~ turn diminishes the 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) + H₂O. (c) The increase of aerosol surface area density (SAD) enhances heterogeneous chemistry in the mid-upper troposphere, reducing the amount of ~~NO_x~~ NO_x and O₃ production and thus of OH. The increased

10
15
20
25

aerosol SAD produces a significant ozone depletion in the stratosphere ~~whose effect is,~~ which results in an increase of UV radiation able to reach the surface: ~~however.~~ However, such effect is overbalanced by the direct scattering of solar radiation, ~~thus in the end the total so that the net~~ amount of tropospheric UV is reduced (except over the polar latitudes) (Pitari et al. (2014); Aquila et al. (2014b)). The high-latitude UV increase has little effect over the methane lifetime, which is mostly influenced from OH changes in the tropics.

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 CH₄ decrease in the upper troposphere. All these effects together produce a longer lifetime of CH₄ that is estimated by the ULAQ-CCM to increase from 8 years for RCP4.5 to 9 years for ~~G4 SG~~ with injection of 5 Tg-SO₂/yr. According to the model, such a lifetime increase is estimated to produce a positive ~~radiative forcing of TOA RF=~~ +0.1 0.11 ± 0.04 W/m² (Pitari et al. (2014); Aquila et al. (2014b)), as an average from year 2020 to 2090.

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 may~~ 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 relative to 2011 of 0.3~~ W/m² (RCP2.6), ~~~2.5-2.2~~ W/m² (RCP4.5), ~~~4-3.7~~ W/m² (RCP6.0), ~~and more than 6.5 and 6.2~~ W/m² (RCP8.5) (Moss et al. (2010) IPCC (2013); 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 SO₂ 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 SO₂ amount and the subsequent AOD is non-linear, as larger amounts of SO₂ will produce larger aerosol particles and the aerosol scattering efficiency decreases. Furthermore, the gravitational 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. (2014a) (Aquila et al. (2014a)). As showed in by Pitari et al. (2016b) 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. (2014a) 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

Figure 4 summarizes the RF values associated with SG found in several published studies breakdown per component, including direct and indirect effects of SG, as discussed in subsections 2.1 and 2.2 and based on published estimates. Aside from the direct effect of sulfate aerosol scattering, only we see that the changes in cirrus-UT ice particle formation and size may produce a RF of comparable magnitude significant negative RF, due to the thermal-dynamical induced thinning of cirrus clouds formed via homogeneous freezing. The indirect effects related to SG-induced changes in GHG concentrations (CH₄, O₃, stratospheric H₂O) are at least approximately 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 Fig. 4, we find that the sum of all possible effects of both 2.5 and direct and indirect RFs of SG with an injection of 5 Tg-SO₂/yr injection results in an overcompensation of the average positive RF over the 2020-2070 period, for the RCP6.0 accounts for $-1.4 \pm 0.4 \text{ W/m}^2$, which means a compensation of the projected positive RF in 2100 relative to 2011 by 64%, 38% and 23% for the IPCC 'realistic' scenarios RCP4.5 cases. A still incomplete average compensation is achieved in the most pessimistic, RCP6.0 and RCP8.5 case, more evident for the 2.5 Tg-SO₂/yr (+0.55, respectively. The November 2015 Paris Agreement aims to strengthen the global response to the threat of climate change by keeping a global temperature rise this century well below 2 °C above pre-industrial levels and to pursue efforts to limit the temperature increase even further to 1.5 °C. According to the IPCC (2013), the best estimate of the total anthropogenic RF relative to 1750 is 2.29 W/m^2):-

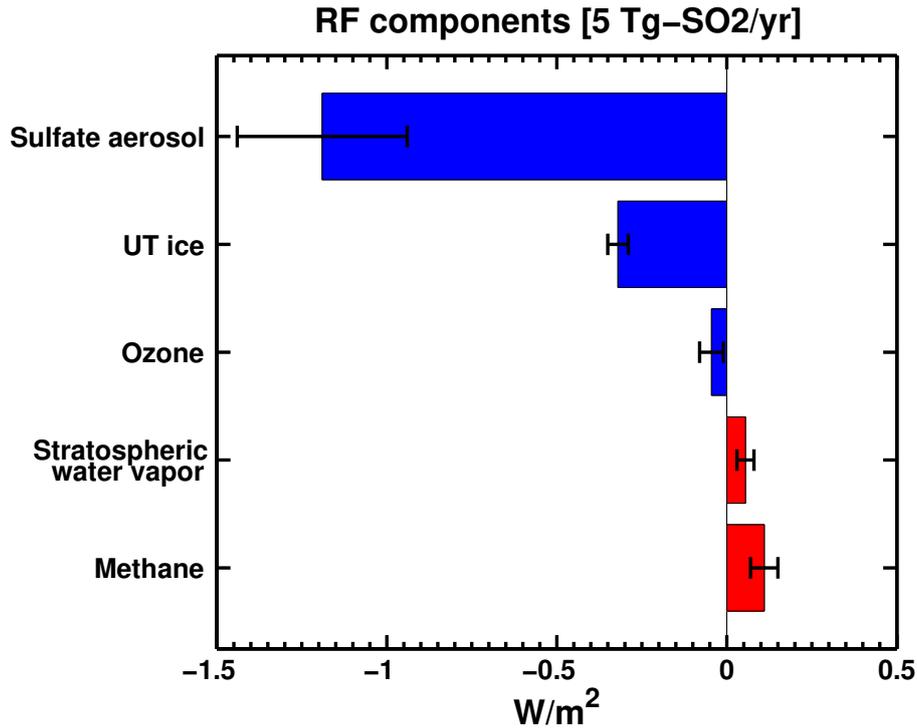


Figure 4. Summary of direct and indirect SG TOA RF per component (see subsections 2.1 - 2.2) (global mean values).

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. (2014) (b), Aquila et al. (2014a) (c), Heckendorn et al. (2009) (d), Niemeier and Timmreck (2015) (e), Kuebbeler et al. (2012) (f), Aquila et al. (2014b) (g). The calculated average stratospheric AOD changes (at $\lambda=0.55 \mu m$) are 0.066 ± 0.026 and 0.045 ± 0.005 for 5 and 2.5 Tg-SO₂/yr injection, respectively. The asterisks denote a first approximation RF estimate, using a linear scaling of the 5 Tg-SO₂/yr value to the 2.5 Tg-SO₂/yr case using the ratio of the stratospheric AODs, when the model calculated RF value is not available for the 2.5 Tg-SO₂/yr case. Mean values and related uncertainties for the SO₄ direct RF are calculated from an average of the available published results. Radiative forcing effects RF (in 2011 and the increase in global mean surface temperature over the period 1880 to 2012 is 0.85 °C. This means that the 2100 RF relative to 2011 projected in the three RCPs (2.2, 3.7 and 6.2 W/m²) RF, respectively) could not allow reaching the Paris Agreement target of a maximum temperature increase of ~0.6 °C up to ~1.1 °C in the period 2011 to 2100. In the hypothesis of SG implementation during the 21st century, with the previously estimated RF = $-1.4 \pm 0.4 W/m^2$ (SG → 5 Tg-SO₂/yr) (SG → 2.5 Tg-SO₂/yr) Baseline, the Paris Agreement target could likely be reached in case of simultaneous WMGHG emissions regulated under scenario RCP4.5 (2020-2070 mean) [WMGHGs+O₃+tropospheric aerosol] Baseline or (barely) under scenario RCP6.0 (2020-2070 mean) [WMGHGs+O₃+tropospheric aerosol] Baseline RCP8.5 (2020-2070 mean) [WMGHGs+O₃+tropospheric aerosol] Sulfate

aerosol scattering -1.45 ± 0.65 (b,c,d,e) -1.15 ± 0.15 (e) 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] $+0.1$ $+0.55$ assuming a climate sensitivity of 0.5 K/Wm^{-2} .

5 3 Conclusions

Our assessment of the published literature on SG concludes that ~~a SG~~ this proposed geoengineering technique has the potential to offset ~~the RF due to GHG, even considering unwanted side effects such as cirrus changes and SG induced increases in~~ a significant part of the positive RF estimated during this century as a consequence of the increasing GHG concentrations. Both direct and indirect effects related to the stratospheric injection of $5 \text{ Tg-SO}_2/\text{yr}$ need to be taken into account to produce robust conclusions. 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.

Previous research works on SG have focused on specific aspects of formation, transport and removal of stratospheric aerosols under geoengineering conditions. However, significant feedback mechanisms exist among the magnitude and location of SO_2 injection, aerosol microphysics, background stratospheric dynamics, aerosol induced changes of SSTs, stratospheric heating rates and large scale circulation. For this reason, designing model simulations in which all these aspects are explicitly linked together is essential for producing more robust estimates of the direct and indirect effects of SG.

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 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~~ endeavour. 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.

Acknowledgements. The authors would like to thank U. Niemeier for helpful discussions.

References

- Aquila, V., Oman, L. D., Stolarski, R., Douglass, A. R., and Newman, P. A.: The Response of Ozone and Nitrogen Dioxide to the Eruption of
5 Mt. Pinatubo at Southern and Northern Midlatitudes, *Journal of the Atmospheric Sciences*, 70, 894–900, doi:10.1175/JAS-D-12-0143.1, <http://dx.doi.org/10.1175/JAS-D-12-0143.1>, 2013.
- Aquila, V., Garfinkel, C., Newman, P., Oman, L., and Waugh, D.: Modifications of the quasi-biennial oscillation by a geoengineering perturbation of the stratospheric aerosol layer, *Geophysical Research Letters*, 41, 1738–1744, 2014a.
- Aquila, V., Pitari, G., Tilmes, S., Cionni, I., De Luca, N., Di Genova, G., and Iachetti, D.: Sensitivity of Methane Lifetime and Transport
10 to Sulfate Geoengineering, in: AGU, San Francisco, Calif., 15-19 Dec, vol. Abstract GC13I-0787, <http://abstractsearch.agu.org/meetings/2014/FM/A23J-3389.html>, 2014b.
- Budyko, M. I.: The Climate of the Future, American Geophysical Union, doi:10.1002/9781118665251.ch7, <http://dx.doi.org/10.1002/9781118665251.ch7>, 2013.
- Canty, T., Mascioli, N. R., Smarte, M. D., and Salawitch, R. J.: An empirical model of global climate ? Part 1: A critical evaluation of
15 volcanic cooling, *Atmospheric Chemistry and Physics*, 13, 3997–4031, doi:10.5194/acp-13-3997-2013, <http://www.atmos-chem-phys.net/13/3997/2013/>, 2013.
- CEC: Climate Engineering Conference 2014, Critical Global Discussion, in: Conference report, Institute for Advanced Sustainability Studies (IASS), Potsdam, http://ce-conference.org/sites/default/files/wysiwyg/files/cec14_conference_report.pdf, 2014.
- Cirisan, A., Spichtinger, P., Luo, B. P., Weisenstein, D. K., Wernli, H., Lohmann, U., and Peter, T.: Microphysical and radiative changes in cirrus clouds by geoengineering the stratosphere, *Journal of Geophysical Research: Atmospheres*, 118, 4533–4548, doi:10.1002/jgrd.50388,
20 <http://dx.doi.org/10.1002/jgrd.50388>, 2013.
- Crutzen, P. J.: Albedo Enhancement by Stratospheric Sulfur Injections: A Contribution to Resolve a Policy Dilemma?, *Climatic Change*, 77, 211–220, doi:10.1007/s10584-006-9101-y, <http://dx.doi.org/10.1007/s10584-006-9101-y>, 2006.
- Dessler, A. E., Schoeberl, M. R., Wang, T., Davis, S. M., and Rosenlof, K. H.: Stratospheric water vapor feedback, *Proceedings of the
25 National Academy of Sciences*, 110, 18 087–18 091, doi:10.1073/pnas.1310344110, <http://www.pnas.org/content/110/45/18087.abstract>, 2013.
- Dutton, E. G. and Christy, J. R.: Solar radiative forcing at selected locations and evidence for global lower tropospheric cooling following the eruptions of El Chichon and Pinatubo, *Geophysical Research Letters*, 19, 2313–2316, doi:10.1029/92GL02495, <http://dx.doi.org/10.1029/92GL02495>, 1992.
- English, J., Toon, O., and Mills, M.: Microphysical simulations of sulfur burdens from stratospheric sulfur geoengineering, *Atmospheric
30 Chemistry and Physics*, 12, 4775–4793, 2012.
- Forster F., P. M. and Shine, K. P.: Stratospheric water vapour changes as a possible contributor to observed stratospheric cooling, *Geophysical Research Letters*, 26, 3309–3312, doi:10.1029/1999GL010487, <http://dx.doi.org/10.1029/1999GL010487>, 1999.
- Gottelman, A., Liu, X., Ghan, S. J., Morrison, H., Park, S., Conley, A. J., Klein, S. A., Boyle, J., Mitchell, D. L., and Li, J.-L. F.: Global
35 simulations of ice nucleation and ice supersaturation with an improved cloud scheme in the Community Atmosphere Model, *Journal of Geophysical Research: Atmospheres*, 115, n/a–n/a, doi:10.1029/2009JD013797, <http://dx.doi.org/10.1029/2009JD013797>, d18216, 2010.
- Heckendorn, P., Weisenstein, D., Fueglistaler, S., Luo, B. P., Rozanov, E., Schraner, M., Thomason, L. W., and Peter, T.: The impact of geoengineering aerosols on stratospheric temperature and ozone, *Environmental Research Letters*, 4, 045 108, <http://stacks.iop.org/1748-9326/4/i=4/a=045108>, 2009.

- Hendricks, J., Karcher, B., and Lohmann, U.: Effects of ice nuclei on cirrus clouds in a global climate model, *Journal of Geophysical Research: Atmospheres*, 116, doi:10.1029/2010JD015302, <http://dx.doi.org/10.1029/2010JD015302>, d18206, 2011.
- 5 IPCC: *Climate Change 2007: The Physical Science Basis*, Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, S. Solomon et al., Cambridge Univ. Press, Cambridge, 2007.
- IPCC: *Climate Change 2013: The Physical Science Basis*. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, T.F. Stocker et al., Cambridge Univ. Press, Cambridge, United Kingdom and New York, NY, USA, 2013.
- 10 Karcher, B. and Lohmann, U.: A parameterization of cirrus cloud formation: Homogeneous freezing of supercooled aerosols, *Journal of Geophysical Research: Atmospheres*, 107, doi:10.1029/2001JD000470, <http://dx.doi.org/10.1029/2001JD000470>, 2002.
- Kravitz, B., Robock, A., Boucher, O., Schmidt, H., Taylor, K. E., Stenchikov, G., and Schulz, M.: The Geoengineering Model Intercomparison Project (GeoMIP), *Atmospheric Science Letters*, 12, 162–167, doi:10.1002/asl.316, <http://dx.doi.org/10.1002/asl.316>, 2011.
- Kravitz, B., Robock, A., and Haywood, J. M.: Progress in climate model simulations of geoengineering, *Eos, Transactions American Geophysical Union*, 93, 340–340, doi:10.1029/2012EO350009, <http://dx.doi.org/10.1029/2012EO350009>, 2012.
- 15 Kravitz, B., Robock, A., and Irvine, P.: Robust Results From Climate Model Simulations of Geoengineering, *Eos, Transactions American Geophysical Union*, 94, 292–292, doi:10.1002/2013EO330005, <http://dx.doi.org/10.1002/2013EO330005>, 2013.
- Krueger, A., Walter, L., Bhartia, P., Schnetzler, C., Krotkov, N., Sprod, I. t., and Bluth, G.: Volcanic sulfur dioxide measurements from the total ozone mapping spectrometer instruments, *Journal of Geophysical Research: Atmospheres*, 100, 14 057–14 076, 1995.
- 20 Kuebbeler, M., Lohmann, U., and Feichter, J.: Effects of stratospheric sulfate aerosol geo-engineering on cirrus clouds, *Geophysical Research Letters*, 39, doi:10.1029/2012GL053797, <http://dx.doi.org/10.1029/2012GL053797>, l23803, 2012.
- Labitzke, K. and McCormick, M. P.: Stratospheric temperature increases due to Pinatubo aerosols, *Geophysical Research Letters*, 19, 207–210, doi:10.1029/91GL02940, <http://dx.doi.org/10.1029/91GL02940>, 1992.
- Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, *Atmospheric Chemistry and Physics*, 5, 715–737, doi:10.5194/acp-5-715-2005, <http://www.atmos-chem-phys.net/5/715/2005/>, 2005.
- 25 Lohmann, U. and Karcher, B.: First interactive simulations of cirrus clouds formed by homogeneous freezing in the ECHAM general circulation model, *Journal of Geophysical Research: Atmospheres*, 107, AAC 8–1–AAC 8–13, doi:10.1029/2001JD000767, <http://dx.doi.org/10.1029/2001JD000767>, 2002.
- McClellan, J., Keith, D. W., and Apt, J.: Cost analysis of stratospheric albedo modification delivery systems, *Environmental Research Letters*, 7, 034 019, <http://stacks.iop.org/1748-9326/7/i=3/a=034019>, 2012.
- 30 McLandress, C., Shepherd, T. G., Jonsson, A. I., von Clarmann, T., and Funke, B.: A method for merging nadir-sounding climate records, with an application to the global-mean stratospheric temperature data sets from SSU and AMSU, *Atmospheric Chemistry and Physics*, 15, 9271–9284, doi:10.5194/acp-15-9271-2015, <http://www.atmos-chem-phys.net/15/9271/2015/>, 2015.
- Meinshausen, M., Smith, S. J., Calvin, K., Daniel, J. S., Kainuma, M. L. T., Lamarque, J.-F., Matsumoto, K., Montzka, S. A., Raper, S. C. B., Riahi, K., Thomson, A., Velders, G. J. M., and van Vuuren, D. P.: The RCP greenhouse gas concentrations and their extensions from 1765 to 2300, *Climatic Change*, 109, 213–241, doi:10.1007/s10584-011-0156-z, <http://dx.doi.org/10.1007/s10584-011-0156-z>, 2011.
- Moriyama, R., Sugiyama, M., Kurosawa, A., Masuda, K., Tsuzuki, K., and Ishimoto, Y.: The cost of stratospheric climate engineering revisited, *Mitigation and Adaptation Strategies for Global Change*, pp. 1–22, doi:10.1007/s11027-016-9723-y, <http://dx.doi.org/10.1007/s11027-016-9723-y>, 2016.

- Moss, R. H., Edmonds, J. A., Hibbard, K. A., Manning, M. R., Rose, S. K., van Vuuren, D. P., Carter, T. R., Emori, S., Kainuma, M., Kram, T., Meehl, G. A., Mitchell, J. F. B., Nakicenovic, N., Riahi, K., Smith, S. J., Stouffer, R. J., Thomson, A. M., Weyant, J. P., and Wilbanks, T. J.: The next generation of scenarios for climate change research and assessment, *Nature*, 463, 747–756, <http://dx.doi.org/10.1038/nature08823>, 2010.
- Neely, R. R., Conley, A. J., Vitt, F., and Lamarque, J.-F.: A consistent prescription of stratospheric aerosol for both radiation and chemistry in the Community Earth System Model (CESM1), *Geoscientific Model Development*, 9, 2459–2470, doi:10.5194/gmd-9-2459-2016, <http://www.geosci-model-dev.net/9/2459/2016/>, 2016.
- Niemeier, U. and Timmreck, C.: What is the limit of climate engineering by stratospheric injection of SO₂?, *Atmospheric Chemistry and Physics*, 15, 9129–9141, doi:10.5194/acp-15-9129-2015, <http://www.atmos-chem-phys.net/15/9129/2015/>, 2015.
- Niemeier, U., Schmidt, H., and Timmreck, C.: The dependency of geoengineered sulfate aerosol on the emission strategy, *Atmospheric Science Letters*, 12, 189–194, doi:10.1002/asl.304, <http://dx.doi.org/10.1002/asl.304>, 2011.
- Niemeier, U., Schmidt, H., Alterskjaer, K., and Kristjansson, J. E.: Solar irradiance reduction via climate engineering: Impact of different techniques on the energy balance and the hydrological cycle, *Journal of Geophysical Research: Atmospheres*, 118, 11,905–11,917, doi:10.1002/2013JD020445, <http://dx.doi.org/10.1002/2013JD020445>, 2013.
- Nordhaus, W. D.: The "Stern Review" on the Economics of Climate Change, Working Paper 12741, National Bureau of Economic Research, doi:10.3386/w12741, <http://www.nber.org/papers/w12741>, 2007.
- Oman, L., Waugh, D. W., Pawson, S., Stolarski, R. S., and Nielsen, J. E.: Understanding the Changes of Stratospheric Water Vapor in Coupled Chemistry–Climate Model Simulations, *Journal of the Atmospheric Sciences*, 65, 3278–3291, doi:10.1175/2008JAS2696.1, <http://dx.doi.org/10.1175/2008JAS2696.1>, 2008.
- Pitari, G.: A Numerical Study of the Possible Perturbation of Stratospheric Dynamics Due to Pinatubo Aerosols: Implications for Tracer Transport, *Journal of the Atmospheric Sciences*, 50, 2443–2461, doi:10.1175/1520-0469(1993)050<2443:ANSOTP>2.0.CO;2, 1993.
- Pitari, G., Aquila, V., Kravitz, B., Robock, A., Watanabe, S., Cionni, I., De Luca, N., Di Genova, G., Mancini, E., and Tilmes, S.: Stratospheric ozone response to sulfate geoengineering: Results from the Geoengineering Model Intercomparison Project (GeoMIP), *Journal of Geophysical Research: Atmospheres*, 119, 2629–2653, 2014.
- Pitari, G., Cionni, I., Di Genova, G., Visioni, D., Gandolfi, I., and Mancini, E.: Impact of Stratospheric Volcanic Aerosols on Age-of-Air and Transport of Long-Lived Species, *Atmosphere*, 7, doi:10.3390/atmos7110149, <http://www.mdpi.com/2073-4433/7/11/149>, 2016a.
- Pitari, G., Di Genova, G., Mancini, E., Visioni, D., Gandolfi, I., and Cionni, I.: Stratospheric Aerosols from Major Volcanic Eruptions: A Composition-Climate Model Study of the Aerosol Cloud Dispersal and e-folding Time, *Atmosphere*, 7, 75, doi:10.3390/atmos7060075, <http://dx.doi.org/10.3390/atmos7060075>, 2016b.
- Pitari, G., Visioni, D., Mancini, E., Cionni, I., Di Genova, G., and Gandolfi, I.: Sulfate Aerosols from Non-Explosive Volcanoes: Chemical-Radiative Effects in the Troposphere and Lower Stratosphere, *Atmosphere*, 7, 85, doi:10.3390/atmos7070085, <http://www.mdpi.com/2073-4433/7/7/85>, 2016c.
- Rasch, P. J., Crutzen, P. J., and Coleman, D. B.: Exploring the geoengineering of climate using stratospheric sulfate aerosols: The role of particle size, *Geophysical Research Letters*, 35, n/a–n/a, doi:10.1029/2007GL032179, <http://dx.doi.org/10.1029/2007GL032179>, 102809, 2008.
- Read, W. G., Froidevaux, L., and Waters, J. W.: Microwave limb sounder measurement of stratospheric SO₂ from the Mt. Pinatubo Volcano, *Geophysical Research Letters*, 20, 1299–1302, doi:10.1029/93GL00831, <http://dx.doi.org/10.1029/93GL00831>, 1993.
- Robock, A. and Mao, J.: The volcanic signal in surface temperature observations, *Journal of Climate*, 8, 1086–1103, 1995.

- Robock, A., Marquardt, A., Kravitz, B., and Stenchikov, G.: Benefits, risks, and costs of stratospheric geoengineering, *Geophysical Research Letters*, 36, n/a–n/a, doi:10.1029/2009GL039209, <http://dx.doi.org/10.1029/2009GL039209>, 119703, 2009.
- 5 Robock, A., Kravitz, B., and Boucher, O.: Standardizing experiments in geoengineering, *Eos, Transactions American Geophysical Union*, 92, 197–197, doi:10.1029/2011EO230008, <http://dx.doi.org/10.1029/2011EO230008>, 2011.
- Sanderson, B. M., O’Neill, B. C., and Tebaldi, C.: What would it take to achieve the Paris temperature targets?, *Geophysical Research Letters*, 43, 7133–7142, doi:10.1002/2016GL069563, <http://dx.doi.org/10.1002/2016GL069563>, 2016GL069563, 2016.
- Sassen, K., Wang, Z., and Liu, D.: Global distribution of cirrus clouds from CloudSat/Cloud-Aerosol lidar and infrared pathfinder satellite
10 observations (CALIPSO) measurements, *Journal of Geophysical Research: Atmospheres*, 113, 2008.
- Storelvmo, T., Kristjansson, J., Muri, H., Pfeiffer, M., Barahona, D., and Nenes, A.: Cirrus cloud seeding has potential to cool climate, *Geophysical Research Letters*, 40, 178–182, 2013.
- Storelvmo, T., Boos, W., and Herger, N.: Cirrus cloud seeding: a climate engineering mechanism with reduced side effects?, *Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, 372, 20140116, 2014.
- 15 Stowe, L. L., Carey, R. M., and Pellegrino, P. P.: Monitoring the Mt. Pinatubo aerosol layer with NOAA/11 AVHRR data, *Geophysical Research Letters*, 19, 159–162, doi:10.1029/91GL02958, <http://dx.doi.org/10.1029/91GL02958>, 1992.
- Strom, J., Strauss, B., Anderson, T., Schrader, F., Heintzenberg, J., and Wendling, P.: In Situ Observations of the Microphysical Properties of Young Cirrus Clouds, *Journal of the Atmospheric Sciences*, 54, 2542–2553, doi:10.1175/1520-0469(1997)054<2542:ISOOTM>2.0.CO;2, [http://dx.doi.org/10.1175/1520-0469\(1997\)054<2542:ISOOTM>2.0.CO;2](http://dx.doi.org/10.1175/1520-0469(1997)054<2542:ISOOTM>2.0.CO;2), 1997.
- 20 Tabazadeh, A., Drdla, K., Schoeberl, M. R., Hamill, P., and Toon, O. B.: Arctic ‘ozone hole’ in a cold volcanic stratosphere, *Proceedings of the National Academy of Sciences*, 99, 2609–2612, doi:10.1073/pnas.052518199, <http://www.pnas.org/content/99/5/2609.abstract>, 2002.
- Taylor, K. E., Stouffer, R. J., and Meehl, G. A.: An overview of CMIP5 and the experiment design, *Bulletin of the American Meteorological Society*, 93, 485, 2012.
- Tilmes, S., Müller, R., and Salawitch, R.: The Sensitivity of Polar Ozone Depletion to Proposed Geoengineering Schemes, *Science*, 320,
25 1201–1204, doi:10.1126/science.1153966, <http://science.sciencemag.org/content/320/5880/1201>, 2008.
- Tilmes, S., Garcia, R. R., Kinnison, D. E., Gettelman, A., and Rasch, P. J.: Impact of geoengineered aerosols on the troposphere and stratosphere, *Journal of Geophysical Research: Atmospheres*, 114, n/a–n/a, doi:10.1029/2008JD011420, <http://dx.doi.org/10.1029/2008JD011420>, d12305, 2009.
- Tilmes, S., Kinnison, D. E., Garcia, R. R., Salawitch, R., Canty, T., Lee-Taylor, J., Madronich, S., and Chance, K.: Impact of very short-lived
30 halogens on stratospheric ozone abundance and UV radiation in a geo-engineered atmosphere, *Atmospheric Chemistry and Physics*, 12, 10945–10955, doi:10.5194/acp-12-10945-2012, <http://www.atmos-chem-phys.net/12/10945/2012/>, 2012.
- Tilmes, S., Sanderson, B. M., and O’Neill, B. C.: Climate impacts of geoengineering in a delayed mitigation scenario, *Geophysical Research Letters*, 43, 8222–8229, doi:10.1002/2016GL070122, <http://dx.doi.org/10.1002/2016GL070122>, 2016GL070122, 2016.
- Trepte, C. R. and Hitchman, M. H.: Tropical stratospheric circulation deduced from satellite aerosol data, *Nature*, 355, 626–628, <http://dx.doi.org/10.1038/355626a0>, 1992.