## Answers to the editor on the ACPD paper (acp-2015-25)

What is the limit of stratospheric sulfur climate engineering?

Ulrike Niemeier and Claudia Timmreck Max Planck Institute for Meteorology, Bundesstr. 53, 20146 Hamburg, Germany

### Dear Dr Kravitz,

I send you a revised manuscript and the answers to the reviewers. We tried to answer the questions carefully and made several changes in the text. Following two reviewers comments on putting more emphasis on different injection heights, we realized that we had not taken into account the increase in the vertical extension of the injections in the English et al (2012) and Pierce et al (2010) paper. We based our arguments on the meridional extension of the injection area only. We included two additional simulations into the paper, with now much closer results to the global values of the other studies. This was for us the basis not to take into account some additional simulations wanted by reviewer 3e.g. additional simulations for 100 Mt(S)/y. We decided that this would broaden the papers topic too strongly.

However, the comments of the reviewers and the thoroughly suggestions help us to improve many parts of the paper, especially the comparison to previous studies. We performed two additional simulations with an increased injection height (24 km) and two different meridional extensions (grid box and 30N to 30S). We added an extra sub-section for this topic, added the two simulations to table 2 and changed the text in the comparison to other model accordingly. Our main conclusion there is: From Geo10-high and Geo10-30-high can we see that the main impact on efficiency is the increase in injection height. Increasing the area in meridional directions decreases the efficiency. Differences between the studies in meridional transport remain. We were not be able to come to a conclusion which of the models gives the better results.

We completely rewrote section 4 and changed the headline to: Limit, uncertainties, and consequences of strong sulfur injections? We included a short discussion of the uncertainties estimated from the experiment design and the model concept. We also include a discussion on some possible impacts: 'What would be the consequences of a 5.5 W m<sup>-2</sup> reduction of the forcing' and discuss briefly impacts on precipitation, ozone, cloud condensation particles, which were estimated from previous studies.

Changes in the text are highlighted in blue. May further changes were done related to the reviewers comment, e.g. the title, and can be seen in the answers to the reviewers.

Thank you very much and with best regards, Ulrike Niemeier

Answers to reviewers on ACPD paper (acp-2015-256)

What is the limit of stratospheric sulfur climate engineering?

Ulrike Niemeier and Claudia Timmreck

Max Planck Institute for Meteorology, Bundesstr. 53, 20146 Hamburg, Germany

We thank all reviewers for the careful reading and the thoroughly suggestions. They help us to improve many parts of the paper, especially the comparison to previous studies. Two reviewers were asking on putting more emphasis onto the injection height. We performed two additional simulations with an increased injection height (24 km) and two different meridional extensions (grid box and 30N to 30S). Efficiency is increased by 50% and 36% in these simulations. This allowed a much better comparison to the results of English et al (2012) and Pierce et al (2010) and helped to explain the differences (Fig. 1). We added an extra sub-section for this topic, added the two simulations to table 2 and changed the text in the comparison to other models accordingly. Our main conclusion there is: From Geo10-high and Geo10-30-high can we see that the main impact on efficiency is the increase in injection height, while the increase of the area in meridional directions decreases the efficiency. We assume this also be valid for the difference between "NARROW" and "BROAD" in E12.

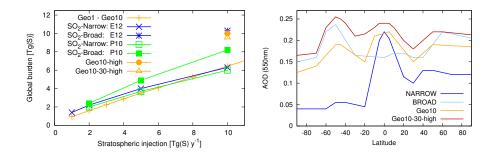


Figure 1: Left: The global sulfate aerosol burden for ECHAM5-HAM simulations Geo1 to Geo10 compared with results from Pierce et al. (2010), P10, and English et al. (2012), E12, for two different emission areas: NARROW, 5 N to 5 S, and BROAD, 30 N to 30 S. In the BROAD simulation the injection area is additionally increased vertically to 20 – 24 km. Right: Plots comparing the zonal mean of the AOD for a narrow and a broad injection area. Plots were created using smoothed values of Geo10 and Geo10-30-high and estimated from "SO<sub>2</sub> NARROW" and "SO<sub>2</sub> BROAD" data after English et al. (2012).

We completely rewrote section 4 and changed the headline to: Limit, uncertainties, and consequences of strong sulfur injections? We included a short discussion of the uncertainties estimated from the experiment design and the model concept. We also include a discussion on some possible impacts: 'What would be the consequences of a 5.5 W m<sup>-2</sup> reduction of the forcing'' and discuss briefly impacts on precipitation, ozone, cloud condensation particles, which were estimated from previous studies.

# Answers to reviewer1 on ACPD paper (acp-2015-256)

## **Specific Comments**

1. Page 10943, lines 5-14: A few words describing how atmospheric oxidants are handled would be useful, i.e. whether they're prescribed or modeled interactively.

We added A simple stratospheric sulfur scheme is employed in model levels at the tropopause and above (Timmreck et al. 2001; Hommel et al. 2011b). The gaseous precursor species (OH, NO2, and O3) are prescribed on a monthly bases as well as photolysis rates of OCS, SO2, H2SO4 SO3, and O3. OCS concentrations are prescribed at the surface and transported within the model.

2. Page 10946, lines 18-20: Aerosol number in the coarse mode also appears to increase more rapidly than that in the accumulation mode (the lines in Fig. 1 are more widely separated in the coarse mode when compared with the accumulation mode), which might be worth mentioning.

We included into the text: As injection rates increase, particle number and radii increase stronger in coarse mode than in accumulation mode.

3. Page 10952, line 21: It would help if it was made clear which of the seven lines in E12's Fig. 9 were used and how they were "estimated and simplified". Was there any scaling of the 525nm AOD in E12's Fig. 9 to the 550nm AOD used here?

The data are not scaled from 523nm to 550nm. We tested the difference between these bands in a radiation simulation and got a difference of 3%. Regarding the overall larger uncertainties we decided not to take this into account in the paper.

The data for the Figure were taken from the Fig. 9 in E12: 'SO2 Broad' and and 'SO2 Narrow'. We do not have the original data. The resulted curve was smoothed to show the main features and not each single maximum and minimum. We change the text to: we draw a schematic diagram of the zonally averaged AOD obtained for a narrow and a broad injection area (Fig., right) after ECHAM5-HAM results and after estimated and smoothed values from Fig. 9 in E12. The curves were smoothed for a better overview.

4. Page 10945, lines 7-11; Page 10953, lines 13-15; Page 10955, lines 1-4: Throughout the paper (I've just selected certain points where the issue is addressed) it needs to be emphasized that the values obtained in this study are for the specific injection altitude chosen. The authors mention (p.10945) that increasing the injection height also increases the efficiency, but there is no quantitative analysis of this effect. This point needs to be made again later in the paper in Sections 4 and 5 where a specific forcing value or values are discussed - these only apply for the altitude chosen. Some quantitative estimate of the range of how the forcing and efficiency might vary with injection height is desirable.

We thank the reviewer to insist on putting more emphasis onto the injection height. We performed two additional simulations with an increased injection height (24 km) and two different meridional extensions (grid box and 30N to 30S). See the text above fore more details.

5. Page 10953, lines 17-24: This Section is the biggest problem with the manuscript. After careful explanation and analysis up to this point the paper loses its way here.

We completely rewrote this section and changed the headline to: "Limit, uncertainties, and consequences of strong sulfur injections". See the text above fore more details.

### Specifically:

(a) What is the source of the "flight emissions" data? Are the emissions comparable to the geoengineering levels under discussion? (line 17).

The values were calculated after the payload given in Robock et al (2008). We skipped this part and discuss only very briefly possible injection heights (after McClellan et al):...Increasing the injection height would reduce the required amount of SO2. However, this would be technically much more challenging. Following McClellan et al. (2012) many existing planes would require technical changes to reach a height of 18 to 20 km. Only Boing F14E may reach higher levels, which otherwise could only be achieved by newly developed technology like hybrid air ships....

(b) I can't make sense of the phrase "injection efficiency given per achieved reduction of TOA forcing in Wm-2" (line 18).

(c) The efficiency of SO2 injection was defined previously (page 10945, lines 20-21) as "the ratio of the top of atmosphere (TOA) forcing to injection strength", which is quite clear. Here, however, it's defined differently as "the amount of sulfur per Wm-2 which is needed to get a certain TOA forcing" (lines 19-20), which I don't follow.

(d) Then (line 20) there's a reference to "These data" - which data?

(e) Then follows (lines 20-22 and more in the next paragraph) some numerical values which appear out of nowhere with no explanation of their source. Neither do I understand what they mean. In lines 20-22 it says "to obtain a reduction of -1 Wm-2 an injection of 4.5 Tg(S)/yr per Wm-2 is necessary, while -7 Wm-2 TOA forcing requires an injection of almost 10Tg(S) per Wm-2". Where do these numbers come from? What does it mean to describe an injection in units of "Tg(S)/yr per Wm-2"? To me, an injection rate is an amount of substance per unit time, so I don't understand what it means to describe an injection in terms of mass per unit time per Wm-2. This section, where the central question of the paper's title is finally addressed, needs to be thoroughly revised. As it stands it makes no sense to this reviewer.

Thank you very much for pointing this out. We revised the text accordingly. The Points b) to e) were taken into account when rewriting the section. The text mentioned in c) and e) is removed.

### **Minor Comments/Technical Corrections**

Comments 1 to 10 were done as suggested. Thank you for the careful corrections.

11. Page 10961, caption to Table 1: Include a few words to make it clear that 'Geo10', which occurs a lot in the text but is not in the Table, is just the 10 Tg(S)/yr version of simulation 'Geo'.

### We added a note to the table caption.

12. Page 10963, Figure 1 (Left): Remove the crosses from the part of the curve for injection rate values greater than 100 Tg(S)/yr: having them on the plot implies that simula-

tions were done for these rates (at about 120, 140, 160, 180 and 200 Tg(S)/yr) but the text suggests that this is just an extrapolation of the fit.

### Done.

13. Page 10964, Figure 2: A color blind person is likely to find this plot difficult to interpret - I suggest either changing to a 'colourblind accessible' set of colours or taking a different approach to this plot.

We tried to improve the figure by changing the colors and reducing the number of lines.

14. Page 10967, caption to Figure 5: The phrase 'with different injection rates of 10 Tg(S)/yr' doesn't really mean anything. If they're all injecting at 10 Tg(S)/yr then the rates are not different. I think you mean that they all inject at the same rate but have different injection strategies or implementations.

Thank you. We changed the caption text.

# Answers to reviewer 2 on the ACPD paper (acp-2015-256)

### **General Comments**

I find the title somewhat misleading. The paper deals with climate engineering by injection of SO2 only. Other methods have been explored to inject sulfur (e.g. H2SO4 by Pierce et al., 2010; and OCS injection) which are not dealt with here, and these could be expected to have different efficiencies and limits. A more appropriate title might be 'What is the limit of climate engineering by stratospheric injection of SO2?'

Thanks for this suggestion for a more precise title. We adopted the suggestion.

This paper represents a contribution to the literature on geoengineering by solar radiation management. However, the main conclusion is not new or surprising. That geoengineering injections become less efficient with increasing emissions has been demonstrated and discussed previously by Heckendorn et al. (2009), English et al. (2012) and Pierce et al. (2010).

This was not meant to be the main conclusion of this paper. This conclusion from previous publications raised the question what happens if we want to counterbalance RCP8.5 forcings? Would this be possible or not, as efficiency decreases. We write in the introduction: With increasing injection rate the forcing efficiency, the ratio of sulfate aerosol forcing to injection rate, decreases Heckendorn et al. (2009). This decrease in forcing efficiency is non-linear and the injected SO2 amount needed to reduce strong GHG forcings will be high. This raised the discussion if it will be possible to counteract strong GHG forcing, like RCP8.5, down to e.g. to a level anticipated for 2020 or not. We try, therefore, to estimate a theoretical upper limit for possible SO2 injections after which a further increase in injection rate causes only a negligible decrease in radiative forcing.

This paper does look at sensitivity to injection region both longitudinally and meridionally, which has been discussed in much less detail by previous authors. It also attempts to derive an upper limit for TOA radiative forcing that could be achieved by SO2 injection, though the uncertainty in this number is large and its utility questionable. And it is the first paper to include radiative feedback in the calculation of aerosol distributions as part of the sensitivity calculations.

# We performed two additional simulations with an increased injection height (24 km) and two different meridional extensions (grid box and 30N to 30S) to further complete this study.

The authors treat the subject of geoengineering by solar radiation management as if there is only one possible method (injection of SO2) and if employed, it would be used to halt future global warming. A more thorough discussion would mention other methods, such as injection of H2SO4 or solid particles, e.g. soot or TiO2. It should also be mentioned that amounts of geoengineering which slow, rather than attempt to halt, surface temperature rise, may have a role in an effective climate strategy (see e.g. MacMartin et al., 2014).

We added a paragraph on other methods to the conclusions: Similar to the injection of SO2 also aerosols could be injected. Ferraro and Charlton-Oerez (2011) studied the impact of limestone, Titania and soot. Soot has a large green house effect, which reduced its efficiency and the simulated forcing of Titania showed strong dependencies on the particle

size, with even positive forcing. Following Weisenstein and Keith (2015) any solid aerosol introduced into the stratosphere would grow via coagulation and accumulation with the consequence of large uncertainties on simulated results. Alternative SRM designs like regional implementation Haywood et al. (2013) or reducing only the rate of temperature increase MacMartin et al. (2015) would require different amounts of SO2 injection in an RCP8.5 scenario.

The fact that the very high levels of geoengineering discussed in this paper would almost certainly present unacceptable risks to ozone gets one brief mention at the end of Section 4. This important point should be included in the introduction as well, and could include an editorial comment that RCP8.5, with continued growth in use of fossil fuels, is extremely undesirable and has no easy fix via geoengineering injection of SO2.

We added some published estimations on ozone impact to the text. The values are very uncertain as all previous studies deal with much smaller amounts of sulfate:Previous geoengineering studies including ozone chemistry estimated changes over the polar region of -10% for an injection of 2 Tg(S)/y (Tilmes et al. (2008)) and around -5% in a multimodel ensemble for 4 to 6 Tg(S)/y (Pitari et al. (2013)). Both studies show a slight increase in the ozone concentration over the Tropics. The impact of higher injection rates might be estimated from volcanic eruptions. Randel et al. (1995) show a decrease of -10% over high latitudes and  $\pm 2\%$  over Tropics from satellite measurements after the Mt.Pinatubo eruption, similar to Aquila et al. (2014). These values seem to be similar to the ones from geoengineering studies. For stronger injection rates only super volcano studies cab be taken as reference. Timmreck and Graf (2006) calculated height dependent changes of + 100% and - 25% in the tropics for a Yellowstone eruption of 850 Tg(S). Bekki et al. (1996) calculated for a simulation of the Toba eruption (about 3000 Tg(S)) a decrease of 40% over the poles and -60% to +150% over the Tropics.

The paper's methodology is sound and generally well documented, with a few exceptions noted under specific comments. The language could use some tweaks to improve the English usage, as noted under 'technical corrections'. In places the discussion could be broadened and I suggest three references to be added. I support publication after the issues detailed here are addressed.

### **Specific Comments**

Page 10941, line 16, and also in Abstract 'These previous studies were performed with SO2 injections of 1 to 10 Tg(S)/yr ' Pierce et al. (2010) show SO2 injections up to 20 MtS/yr. We revised this to up to 20 MtS/yr.

Page 10943, line 7 'Nucleation was adapted to high SO2 concentrations...' Do you mean high H2SO4 concentrations? SO2 plays no direct role in nucleation.

We changed this in the text.

Page 10945, lines 24-25' These data are derived from a double radiation call' Please explain 'double radiation call'

We changed the text to: These data are derived from calling the radiation calculation in the model twice, once without and once with aerosols, whereby only the latter is seen from

the climate model.. With this method we are able to calculate the instantaneous aerosol forcing only and get the radiative forcing of the aerosol.

Page 10951, line 23: The statement that 'the aerosol is coupled to a radiation scheme' in the Pierce et al. (2010) work is misleading. That study and the Heckendorn et al. (2009) study calculated changes in radiation due to aerosols but there was no feedback of radiation into the aerosol distribution. Aerosols were calculated in a 2-D model with fixed circulation off-line from radiative effects.

We took this into account and changed the formulation in the text.

Page 10953-10954, the paragraph spanning this page transition Robock (2009) did not perform a serious analysis of delivery systems for geoengineering. The work of McClellan et al. (2012) covers this topic in more depth and would provide a more appropriate citation. Table 1 of that reference gives number of aircraft required to lift 1 Mt-S per year into the stratosphere for several existing aircraft types. From this, the fleet size required to inject 26 or 45 Tg-S/yr can be estimated.

We rewrote the section and skipped, related to other reviewers comments, the discussion on a necessary amount of flights. But we used the McClellan et al. (2012) paper for citing the possible flight height.

Page 10954, lines 16 - 17 In the discussion of possible cloud feedback, add reference to Cirisan et al. (2013). The reference indicates that this feedback might go either way. I suggest changing 'would' to 'might': 'the resultant brighter clouds might reflect more sunlight, a positive feedback' You might consider replacing 'R TOA'with ' $\Delta$ R TOA' to be clear that you are talking about the change in top-of-atmosphere radiative forcing due to geoengineering aerosols, not total radiative forcing.

We use  $\Delta R$  TOA now and changed the text on cloud feedback to: The resultant brighter clouds might reflect more sunlight, a positive feedback. Cirisan et al (2013) describe the mid-latitudinal averages in the range of  $\pm$  0.04 W m<sup>-2</sup> for injection rates up to 5 Tg(S) yr<sup>-1</sup>. Locally this values can be larger but the global impact can be assumed as small also for larger injection rates. Furthermore Kuebbeler et al. (2012) showed that a vertical shift of the tropopause height caused by the warmer lower stratosphere has implications on cirrus clouds and the cloud top height, with further impacts on the hydrological cycle.

We included the technical correction in the text. Thank you very much for the careful reading and the suggestions.

# Answers to reviewer 3 on the ACPD paper (acp-2015-25)

### **General comments**

Stratospheric dynamics have a critical impact on aerosol microphysics and lifetime. As the authors noted, there is significant disagreement between their broad (30S-30N) injection results and those of Pierce et al. (2010) and English et al (2012), and they speculate that differences in the tropical transport barrier may be a primary reason why. Are there observations or other studies that estimate what the actual transport efficiency across this tropical transport barrier might be? Which model(s) are more accurate - Pierce, English, or this work?

The results of the two additional simulations made it possible to better distinguish between the impact of an increased injection height and an increased meridional extension. The global burden results of the new simulations are now quite close to the ones given in English et al (2012). However we still see differences in the simulated meridional transport between both models. We know from ECHAM5-HAM that the model slightly overestimates the meridional transport. An answer which of the other models is more accurate is quite difficult, most probably better with a detailed model data intercomparison. Here we have to refer to the upcoming SSIRC model intercomparison http://www.sparc-ssirc.org/.

What does the absence of QBO in their model do to the stratospheric circulation? what are the possible errors that arise from it?

Punge et al. (2009) compare zonal mean values of stratospheric CH4 concentrations between the east and west phase of a QBO. Concentrations in the tropics are increased by 10% and decreased by 10 to 15% in extra tropics in an easterly phase at 10 hPa. They state differences in the strength of the transport barrier with QBO phase as reason. Differences in meridional transport were already found by Plumb and Bell (1982). Hommel et al. (2015) found modulations of the size distribution of the aerosol by the QBO but in the bulk of the stratospheric aerosol layer for most of the analyzed parameters (incl. the effective radius) only moderate statistically significant QBO signatures (<10 %). See also later comment on Sec. 2.1

What is the stratospheric age-of-air in their model compared to a best-guess from observations? the ECHAM model has a rather coarse vertical resolution (39 vertical levels); how might that affect stratospheric dynamics and strat-trop exchange?

Based on these dynamical uncertainties, what are estimates regarding how this may translate to errors in geoengineering efficacy? For example, if the age-of-air in your model is 10% too short, does that translate into a geoengineering AOD that is 10% too low? or vice versa.

Age of air (AoA) measures the mean transit time of air parcels along the Brewer-Dobson circulation (BDC) starting from their entry into the stratosphere. AoA is determined both by transport along the residual circulation and by two-way mass exchange (mixing) (Garny et al, 2014). We cannot estimate the age of air from our simulations but previous studies showed an age of air of 3 to 3.5 years in ECHAM6, depending on the model vertical resolution, 4 years in MaEcham5 (Manzini and Feichter (1999)) compared to 4.5 to 5.5 years in measurements (Bunzel and Schmidt (2013)). The comparison of L47 and L95 simula-

tions in Bunzel and Schmidt (2013) shows a smaller age of air for L47 and a slightly higher upward mass flux through the 70-hPa pressure surface. In a simulation with high vertical resolution the QBO is resolved with additional implication on meridional transport (see below).

Garcia and Randel (2008) show an age of air of about 3 years for the WACCM model and values in-between 2.8 and 3.8 years depending on the method (Garcia et al, 2011). Compared to the measurements both models show a too low AoA and we will not gain an answer on the sulfate transport from this value.

New model simulations of 100 Tg injections with modified dynamics (QBO, gravity waves, etc) that alter transport efficiency across tropical barrier and/or stratospheric age-of-air would help quantify these uncertainties.

We agree on that but a detailed investigation of stratospheric transport dynamics could be a topic on its own. Our intention is to explore the limits of climate engineering by stratospheric injection of SO2 and we choose a continuous 10TgS/y as our reference standard case from which we explore the parameter space.

2)Weisenstein et al. (2007) investigated coarse mode widths between 1.45 and 1.58 and found that modal models were accu- rate sometimes, but not always, and there was no single mode width specification that was consistently most accurate. English et al. (2013) calculated equivalent lognormal mode widths from their sectional model after large volcanic eruptions and found the coarse mode widths to vary between 1.2 and 2.0. (Please cite both of these papers). Also, as aerosol size evolves, mode widths can change. 2-moment modal models such as what the authors use here are unable to represent this. Some of these things may be able to be calculated, but others may require new simulations, such as changing the GEO mode width from 1.2 to 2.0 and comparing 100-Tg injections. (my understanding is that the VOLC simulations completed actually remove the coarse mode rather than changing the mode width)

We added to section 3.2.3: Weisenstein et al (2007) compared a modal aerosol model with a fine bin model, showing that with optimized mode width a modal model can describe the distribution of a bin model reasonable. English et al (2013) highlighted the changing mode width over time after a volcanic eruption. This changing time factor is not important under SRM. However, the result show that under different injection rates the mode distribution differs which might alter the TOA radiative forcing as well. We agree with the reviewer that a modal model is a simplification, as all ready stated in the paper. Regarding the uncertainties related to injection area, transport and the fact that the difference between experiment Geo100 and Volc100 is about 6% in TOA forcing, we decided not to put our focus on a sophisticated system of different mode width choice depending on the SO2 concentration. Of course, the choice of the mode width has an impact on the TOA forcing, because the it depends on the particle size(e,g Timmreck et al. (2010), but a detailed discussion of this aspect is beyond the focus of our paper).

3) The authors note the impact of injection height and pulsed injections. At 100 Tg injection rates, how much more effective is an injection at 25 km compared to 19km? At 100 Tg injection rates, how much more effective is a pulsed injection compared to a continuous

injection? New model simulations may be required to confidently include these parameters when calculating an uncertainty range. Based on these additional simulations, and other estimates of uncertainties based on your own calculations and other papers, calculate an uncertainty range of injection rates required to counteract "business as usual" and include that range in the abstract (e.g. maybe it's 20-50 Tg/yr instead of 45 Tg/yr).

Thank you for insisting on further details on the impact of the injection height (details see above). WE also included a value on the impact of pulsed injections and included a summary on the estimated uncertainties into Sect. 4 (details see above). We changed the last sentence in the abstract to: This result implies that the solar radiation management strategy required to keep temperatures constant at that anticipated for 2020, whilst maintaining "business as usual" conditions, would require atmospheric injections into a height of 60 hPa of the order of 45 Tg(S) yr<sup>-1</sup> ( $\pm$  12% or 6 Tg(S) yr<sup>-1</sup>) which amounts to 5 to 7 times that emitted from of the Mt. Pinatubo eruption each year. We rewrote Section 4 and included many of the calculated uncertainties there and additionally into the previous sections.

## Specific suggestions

Title: Either change title to "What is the limit of climate engineering via continuous SO2 injections at 19km altitude", or preferably, conduct more detailed assessment of uncertainty ranges via sensitivity studies, some of which are outlined above.

The title changed slightly to: "What is the limit of climate engineering by stratospheric injection of SO2?" We added simulations with an increased injection height.

Abstract: you use the term "injection strengths" but a more accurate term would be "injection rates". Please go through the manuscript and be consistent with whatever term you decide on. You also use "injection flux" and "emission strength" in other places. I think rate is better than strength or flux.

### Thank you very much for the suggestion. We corrected this in the text.

Abstract: Mention that the 45 TgS/yr calculation comes from continuous so2 injections in a single grid box at 19km altitude, and add uncertainty ranges around it based on the sensitivity studies completed as per my primary suggestions. for example, is your best guess 30 to 60 TgS/yr so2 injection based on uncertainty analysis of stratospheric dynamics, aerosol microphysics, injection domain, etc.

See above.

Section 2.1: What is the chemistry scheme in your model? Please provide this information in the paper revision and a brief citation to or explanation of the pros/cons/possible errors involved with the chemistry scheme on geoengineering efficacy.

We added A simple stratospheric sulfur scheme is employed in model levels at the tropopause and above (Timmreck (2001); Hommel et al. (2011)). The gaseous precursor species (OH, NO2, and O3) are prescribed on a monthly bases, as well as photolysis rates of OCS, H2SO4, SO2, SO3, and O3. OCS concentrations are prescribed at the surface and transported within the model. to the text in the model description and added to the introduction: The model is not coupled to an ocean model, nor is a full atmospheric chemistry module integrated. Thus, impacts on climate or ozone concentrations cannot be

simulated. In Section 4 some estimates after previous studies on a possible impact on the ozone concentration are given.

Section 2.1 para 5: Please clarify how you changed sigma to 1.2 instead of 2; are those results published somewhere?

The sigma value is given as a parameter in the model code. We slightly changed the text in this paragraph. The results are not published previously but given in Figure 1 in this paper. Reason for the change from the set up of Niemeier et al. (2009) is that we used Heckendorn et al. (2009) as reference for our mode setup and decided to stick closer to the setup which was determined in the box model comparison in Kokkola et al. (2009) for the related SO<sub>2</sub> concentration.

Section 2.1 para 6: It seems like QBO could significantly impact your conclusions. What is your rationale for saying that it wouldn't? What is your best guess as to what the AOD and burdens would be if your model did resolve QBO? Would efficacy be better, worse, and/or what is the uncertainty?

The following text was added to Section 3.2.4: This study was performed with a relative coarse vertical resolution of 39 levels up to 0.01 hPa. Increasing the amount of vertical levels and consequently reducing the vertical grid space would slightly increase efficiency due to less numerical diffusion (3% higher burden estimated from a volcanic eruption study). Including the QBO via nudging may also increase efficiency. Punge et al (2009) show that methane concentrations in the tropics change by  $\pm$  10% and by 10 to 15% in extra tropics depending on the QBO phase. These differences are caused by the different meridional transport as a consequence of different stratospheric transport barrier strength between QBO east and west phases Plumb and Bell (1982). A detailed analysis of the QBO impact on the tropical stratospheric aerosol layer was recently published by Hommel et al. (2015). They found in the bulk of the stratospheric aerosol layer for most of the analyzed parameters (incl. the effective radius) only moderate statistically significant QBO signatures (<10 %). Simulating an internally generated QBO like oscillation by increasing the vertical resolution to 90 levels would cause a slowing of the QBO oscillation and for injection rates roughly about 8 Tg(S) yr<sup>-1</sup> a constant QBO west phase in the lower stratosphere with overlaying easterlies. Aquila et al. (2014). Increasing injection rates strengthen the constant QBO west phase and, following Plumb and Bell (1982), decrease efficiency further by reducing the meridional transport.

Section 2.2: you mention other studies that found improved results with increased injection height and pulsed injections. These are important "pieces of the puzzle" for determining what the actual geoengineering limitations might be. See above.

Section 3.1 para 2: In the paragraph starting with "A more detailed illustration.." there are several sentence fragments that could be improved.

Thank you, we changed the text slightly.

Section 3.2.1: The bulleted list 1-4 has several grammatical errors: (improvements suggested): 1. "Nucleation continuously forms new small particles within the injection area." 3. "Due to advection, larger particles in the the accumulation and coarse modes are globally dispersed." 4. "The larger the ratio, the larger the coagulation coefficient." Thank you, we followed this suggestion.

Section 3.2.2 para 1: Yes, you mention the possible impacts of QBO. It would be interesting to do a sensitivity study on the effects of QBO on geoengineering efficacy. At aminimum, estimate the uncertainty in your results based on this.

We refer again to the very detailed study by Hommel et al (2015) who investigate the impact of the QBO on the tropical aerosol layer during volcanically undisturbed times. They found below 10 hPa, in those regions where the aerosol mixing ratio is largest (50–20 hPa, or 20–26 km), that in most of the analyzed parameters only moderate statistically significant QBO signatures (<10 %). This is also valid for the effective radius, where QBO-induced modulations are smaller than 5 %. A detailed sensitivity study would be beyond the scope of the paper. For the changed text see our comment above.

Section 3.3 para 3: What do the observations say about meridional transport/tropical transport barrier? Which of the three models is most accurate? How do these varying results contribute to an uncertainty analysis of the actual limits with stratospheric so2 geo-engineering?

Observations on sulfate transport are only available from short periods after volcanic eruptions, mostly Mt. Pinatubo eruptions. AS 6 weeks later Cerro Hudson erupted, measurements are influenced by a small degree by this eruption. This allows many interpretations of the sulfur transport after the Mt.Pinatubo eruption. Therefore, these questions cannot be answered within this study. The planed intercomparison study within SSIRC may give an answer.

Section 3.3 para 3: It is "AOD", not ADO Done

Section 4 para 2: It would be interesting to calculate the CO2 emissions from 6 million aircraft flights per year. The net geoengineering efficacy would be reduced further due to the LW absorption from additional CO2.

The number of necessary flights are no longer in the text. See also comments to the other reviewers.

Section 4 para 3: grammar error here: " may get via sedimentation...". And after "changes in precipitation" add ", etc." or equivalent.

Section 4 has changed (see above).

Conclusions para 2: grammar: "This study contributes". grammar: "less evenly distributed".

### Done

Conclusions: Here and elsewhere, change "injection flux" to "injection rate" everywhere in the paper.

### We followed this suggestion.

Table 1: Instead of "geoeng" or "volc", it would be more useful to state the mode peaks and widths. Perhaps you could put "geoeng" or "volc" in parantheses.

We added the sigma values to Table 1.

Fig.1: There are specific definitions of "TOA radiative forcing"; please make sure you are consistent with them.

We changed the text to: These data are derived from calling the radiation calculation in the model twice, once without and once with aerosols. With this method we calculate the instantaneous aerosol forcing only and get the radiative forcing of the aerosol.

Fig.2: The legend overlaps with some of the curves, and the y-axis units needs a superscript.

We corrected the legend and slightly changed the figure regarding a comment of reviewer 1.Superscript still missing

Fig.3: First sentence is not clear. Do you mean to say "injected in a one grid box wide area"

We corrected this to: Burden of (left) SO2 and (right) sulfate coarse mode particles as calculated the first grid box along the Equator for two different simulations.

## References

- Aquila, V., Garfinkel, C. I., Newman, P., Oman, L. D., and Waugh, D. W.: Modifications of the quasi-biennial oscillation by a geoengineering perturbation of the stratospheric aerosol layer, Geophys. Res. Lett., 41, 1738–1744, doi:10.1002/2013GL058818, 2014.
- Bekki, S., Pyle, J. A., Zhong, W., Haigh, R. T. J. D., and Pyle, D. M.: The role of microphysical and chemical processes in prolonging the climate forcing of the Toba eruption, Geophys. Res. Lett., 23, 2669–2672, 1996.
- Bunzel, F. and Schmidt, H.: The Brewer-Dobson Circulation in a Changing Climate: Impact of the Model Configuration, J. Atmos. Sci., 70, 1437–1455, doi: http://dx.doi.org/10.1175/JAS-D-12-0215.1, 2013.
- 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, J. Geophys. Res. Atmos., 118, 4533–4548, doi:10.1002/jgrd.50388, 2013.
- English, J. M., Toon, O. B., and Mills, M. J.: Microphysical simulations of sulfur burdens from stratospheric sulfur geoengineering, Atmos. Chem. Phys., 12, 4775–4793, doi: 10.5194/acp-12-4775-2012, 2012.
- English, J. M., Toon, O., and M.J., M.: Microphysical simulations of large volcanic eruptions: Pinatubo and Toba, J. Geophys. Res. Atmos., 118, 1880–1895, doi:10.1002/jgrd.50196, 2013.
- Ferraro, A. J. a. E. H. and Charlton-Oerez, A.: Stratospheric heating by potential geoengineering aerosols, Geophys. Res. Lett., 38, L24 706, doi:10.1029/2011GL049761, 2011.
- Haywood, J., Jones, A., Bellouin, N., and Stephenson, D.: Asymmetric forcing from stratospheric aerosols impacts Sahelian rainfall, Nature Climate Change, 3, 660–665, doi: 10.1038/nclimate1857, 2013.

- 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, Environ. Res. Lett., 4, 045108, doi:10.1088/1748-9326/4/4/045108, 2009.
- Hommel, R., Timmreck, C., and Graf, H. F.: The global middle-atmosphere aerosol model MAECHAM5-SAM2: comparison with satellite and in-situ observations, Geoscientific Model Development, 4, 809–834, doi:10.5194/gmd-4-809-2011, URL http://www.geosci-model-dev.net/4/809/2011/, 2011.
- Hommel, R., Timmreck, C., Giorgetta, M., and Graf, H.: Quasi-biennial oscillation of the tropical stratospheric aerosol layer, Atmos. Chem. Phys., 15, 5557–5584, doi: 10.5194/acp-15-5557-2015, 2015.
- Kokkola, H., Hommel, R., Kazil, J., Niemeier, U., Partanen, A.-I., Feichter, J., and Timmreck, C.: Aerosol microphysics modules in the framework of the ECHAM5 climate model – intercomparison under stratospheric conditions, Geoscientific Model Development, 2, 97–112, URL http://www.geosci-model-dev.net/2/97/2009/, 2009.
- Kuebbeler, M., Lohmann, U., and Feichter, J.: Effects of stratopheric sulfate aerosol geo-engineering on cirrus clouds, Geophys. Res. Lett., 39, L23803, doi: 10.1029/2012GL053797, 2012.
- MacMartin, D., Caldeira, K., and Keith, D.: Solar geoengineering to limit the rate of temperature change, Phil. Trans. R. Soc. A, 372, 20140134, doi:10.1098/rsta.2014.0134, 2015.
- Manzini, E. and Feichter, J.: Simulation of the SF6 tracer with the middle atmosphere MAECHAM4 model: Aspects of the large-scale transport, J. Geophys. Res., 104(D24), 31 097–31 108, doi:10.1029/1999JD900963, 1999.
- McClellan, J., Keith, D. W., and Apt, J.: Cost analysis of stratospheric albedo modification delivery systems, Environmental Research Letters, 7(3), doi:10.1088/748-9326/7/3/034019, 2012.
- Niemeier, U., Timmreck, C., Graf, H.-F., Kinne, S., Rast, S., and Self, S.: Initial fate of fine ash and sulfur from large volcanic eruptions, Atmospheric Chemistry and Physics, 9, 9043–9057, URL http://www.atmos-chem-phys.net/9/9043/2009/, 2009.
- Pierce, J. R., Weisenstein, D. K., Heckendorn, P., Peter, T., and Keith, D. W.: Efficient formation of stratospheric aerosol for climate engineering by emission of condensible vapor from aircraft, GRL, 37, L18805, doi:10.1029/2010GL043975, 2010.
- Pitari, G., Aquila, V., Kravitz, B., Robock, A., Watanabe, S., Luca, N. D., Genova, G. D., Mancini, E., Tilmes, S., and Cionni, I.: Stratospheric ozone response to sulfate geoengineering: Results from the Geoengineering Model Intercomparison Project (GeoMIP), Journal of Geophysical Research, 119, 2629–2653, doi:10.1002/2013JD020566, 2013.

- Plumb, R. A. and Bell, R. C.: A model of quasibiennial oscillation on an equatorial betaplane, Q. J. R. Meteorol. Soc., 108, 335–352, 1982.
- Punge, H. J., Konopka, P., Giorgetta, M. A., and Müller, R.: Effects of the quasi-biennial oscillation on low-latitude transport in the stratosphere derived from trajectory calculations, J. Geophys. Res., 114, D03 102, doi:10.1029/2008JD010518, 2009.
- Randel, W. J., Wu, F., III, J. M. R., Waters, J. W., and Froidevaux, L.: Ozone and temperature changes in the stratosphere following the eruption of Mount Pinatubo, J. Geophys. Res., 100(D8), 16753–16764, doi:10.1029/95JD01001, 1995.
- Tilmes, S., Müller, R., and Salawitch, R.: The sensitivity of polar ozone depletion to proposed geoengineering schemes, Science, 320(5880), 1201–1204, doi: 10.1126/science.1153966, 2008.
- Timmreck, C.: Three–dimensional simulation of stratospheric background aerosol: First results of a multiannual general circulation model simulation, J. Geophys. Res., 106, 28313–28332, 2001.
- Timmreck, C. and Graf, H.-F.: The initial dispersal and radiative forcing of a Northern Hemisphere mid-latitude super volcano: A model study, Atmos. Chem. Phys., 6, 35–49, URL http://www.atmos-chem-phys.net/6/35/2006/, 2006.
- Timmreck, C., Graf, H.-F., Lorenz, S. J., Niemeier, U., Zanchettin, D., Matei, D., Jungclaus, J. H., and Crowley, T. J.: Aerosol size confines climate response to volcanic supereruptions, Geophys. Res. Lett., 37, L2470, doi:10.1029/2010GL045464, 2010.
- Weisenstein, D. K. and Keith, D.: Solar geoengineering using solid aerosol in the stratosphere, Atmos. Chem. Phys. Discuss., 15, 11799–11851, doi:10.5194/acpd-15-11799-2015, 2015.

Weisenstein, D. K., Penner, J. E., Herzog, M., and Liu, X.: Global 2–D intercomparison of sectional and modal aerosol modules, Atmos. Chem. Phys., 7, 2339–2355, 2007.
Garcia, R. R. and W.J. Randel, 2008: Acceleration of the Brewer–Dobson Circulation due to Increases in Greenhouse Gases. J. Atmos. Sci., 65, 2731–2739.doi: http://dx.doi.org/10.1175/2008JAS

Garcia, R. R. and W.J. Randel and D. E. Kinnison, 2011: On the Determination of Age of Air Trends from Atmospheric Trace Species. J. Atmos. Sci., 68, 139–154. doi: http://dx.doi.org/10.1175/2010JAS3527.1

Garny, H., T. Birner, H. Bönisch, and F. Bunzel (2014), The effects of mixing on age of air, J. Geophys. Res. Atmos., 119, 7015–7034, doi:10.1002/2013JD021417.