

Review of “*Exploring accumulation-mode-H₂SO₄ versus SO₂ stratospheric sulfate geoengineering in a sectional aerosol-chemistry-climate model*” by S. Vattioni et al.

Comments by anonymous reviewer #1 are in bold. Author responses are in blue.

This article is an interesting comparison between different sulfate geoengineering (SG) injection strategies, namely between SO₂ injection (the standard for most simulations up to now) and H₂SO₄ (that has been proposed but not as often studied). The study is innovative in its comparison of the two injection strategies (not in the coupling of CCM and sectional aerosol model, as mentioned by the authors at the end of section 2), and I believe that it deserves publication in ACP after the two major issues (and several minor suggestions), that I list below, have been addressed.

We would like to thank reviewer #1 for her/his insightful comments and suggestions. Our point-by-point replies are given below:

Major points:

1) The authors compare their results with previously obtained results (some of them at least), but they have one major difference with them that is not highlighted anywhere: the longitudinal distribution of the emitted sulfate. Let’s take first Niemeier and Timmreck (2015) that the authors mention: they only have one simulation (Geo10-lon) where the injection is spread on all longitudes, and only for a very small latitudinal range (3N to Eq.). In all other scenarios, emissions are only between 120.9 to 123.75 E. Also, Kleinschmitt et al. (2018): they mostly always inject at one single longitude (120E), and only once (BROAD) at 28 locations between 30N and 30S.

So, when the authors in the conclusion of their study claim that their RF efficiencies are smaller than those previous ones and they generally point at “lower stratospheric aerosol burdens in our model” they should explain this major difference.

I’m not suggesting the authors change their simulations to align closer to previous ones, but the difference between their study and previous ones should be further highlighted, and maybe compared with their sensitivity test (GEO_AERO_point_00 and GEO_SO2_point_00) if they want to make an apple to apple comparison. Overall, I think it would be good that the authors justified much better their assumption that injections over all longitudes are preferable to injections centered in one longitude (and so they chose to do pointed injections only as sensitivity case). Do the authors think this is model dependant? For instance, Tilmes et al (2017) note that, in their model, “Single grid point injections produce sulfate aerosols of smaller size that reflect sunlight more efficiently than injections over a longitude band”.

We agree that many other studies have used point injections at one equatorial grid box. Initially, the focus of our study was to investigate differences of emitting SO₂-gas and AM-H₂SO₄ as well as the effect of partially emitting outside the tropical pipe. The distribution over all longitudes was mainly chosen to compare our results to Heckendorn et al. (2009), who used the 2D model AER offline and put the results into SOCOL. However, it turned out that the comparison with Heckendorn et al. (2009) is limited as too many parts of the model have changed since their study.

There have been other studies which investigated injections at all longitudes with point emissions only as sensitivity runs (i.e. English et al., 2012, Pierce et al. 2010). As in our study, English et al. (2012) found only small differences between point and longitudinally spread emissions. So far, there are only few studies which investigated pulsed emissions. The opposing behaviour between SO₂ and AM-H₂SO₄ emission scenarios for sensitivities to temporal and spatial concentrated emissions was first shown in this study.

In our conclusions we changed the scenarios we refer to. We now compare the point emission scenarios to results of Niemeier et al. (2011) and Kleinschmitt et al. (2018). In the rest of the paper we point now to the differences in emission scenarios wherever necessary.

2) In Table 3 they show what they call “important impacts”, naming water vapor and ozone column. However, recent literature has shown that those are not the only (nor the more important) impacts, especially in terms of radiative forcing: what about methane (Visioni et al., 2017 and Tilmes et al., 2018) or ice clouds (Kuebbeler et al., 2012 and Visioni et al., 2018a)? I believe that both factors can’t be ignored, when talking about the possible range of impacts of SG. At the very least they should be mentioned, acknowledging previous results, but much better would be to show the changes produced in SOCOL regarding one or both of these aspects. Considering the authors focus also on chemical changes by showing OH and H₂O vertical profiles, I suggest they at least analyse their atmospheric methane changes, comparing them with previous studies.

There are other side effects of SSG such as tropospheric methane lifetime increase. In our simulation, tropospheric methane mixing ratios remain largely unchanged as we use prescribed mixing ratio boundary conditions for methane at the ground as well as prescribed SST (see figure 1 below). However, the reaction of methane with OH is strongly temperature dependent. In our model we observe a tropospheric temperature decrease of up to 0.95 K which leads to an increase in methane lifetime of up to 2.3 %, while OH concentration almost unchanged in our simulations. Therefore, our model shows a similar effect as in Visioni et al. (2017), but much smaller, as we emitted only 1.83 Mt S per year and Visioni et al. (2017) emitted 5 Mt S per year. When we scale our results linearly to 5 Mt S per year, methane lifetime increases by up to 6.3 % depending on the scenario. This is still less than the 10 % found by Visioni et al. (2017), probably due to the constant SST in our model setup. When taking interactive SST into account, increased changes in temperature, tropospheric ozone and O¹(D) chemistry as well as H₂O concentrations could account for the remaining difference to Visioni et al. (2017). In our simulations, the lifetime of methane at 50 hPa in the lower stratosphere decreases about 14 % in continuous AM-H₂SO₄ emission scenarios at all longitudes and about 10 % in the corresponding SO₂ emission scenarios, which is in agreement with the OH changes described above.

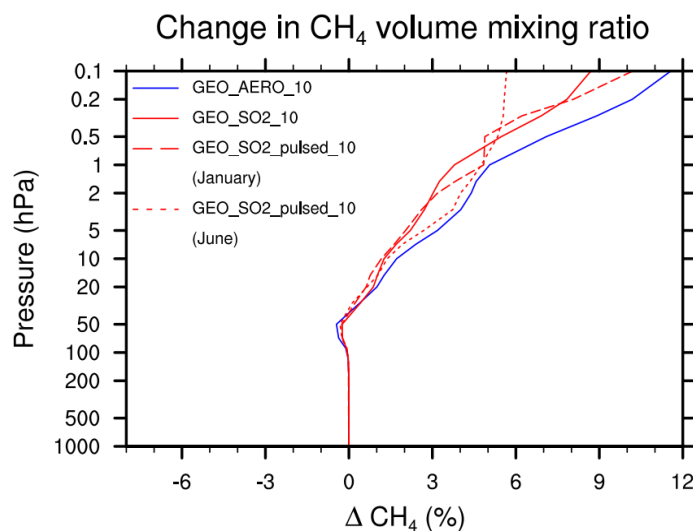


Figure 1. Vertical profile of the methane volume mixing ratio anomaly. Anomalies represent difference compared to the background simulation. Data show annual and zonal averages between 15° N and 15° S except where indicated. The figure shows results for injections within 10° of the equator as well as zonal averages of January (emissions during January 1st and 2nd) and June (month before emission) from the pulsed simulation.

Effects on ice clouds cannot be investigated with SOCOL-AER since the model does not include a comprehensive cloud microphysics scheme.

We now clearly state in the text that there are further potential side effects which are not investigated in this study, and discuss the results of the studies mentioned by the referee. We further clarify that Table 3 refers to the negative side effects investigated in this study.

As I said, before being suitable for publication, I believe a major revision considering these two points is necessary. Furthermore, I have some minor points that I list below:

Abstract: The abstract is way too long and confusing. You don't really need to put all your results in the abstract, there's plenty of space elsewhere. Also, the use of the term "surprising" twice is misleading, considering that the last phrase of the abstract is "this study corroborates previous studies". Shorten it by only pointing at some of the results and leave for the discussion all the rest. I agree with Alan Robock that the usage of parenthesis in that way needs to be dropped.

The abstract has been rewritten and shortened. We now focus on the most important conclusions of the paper, removed the use of the word "surprising" and avoided the overly use of parenthesis. We also adapted the last sentence of the abstract as the part "this study corroborates previous studies" was misleading and weakens new findings made in this paper.

P. 1, lines 17-19: This phrase is very convoluted and confusing.

This sentence has been changed.

P. 2, lines 15-25: As I mentioned in my point 2), are the authors referring to only SG side-effects that reduce the efficiency of the RF produced by the sulfate aerosols (as stated in lines 17-18) or general drawback of SG? I believe the former would be more correct and would make more sense here. Because of this, a bit of clarification is important: (2) of all chemical side effects, ozone depletion is the one with the less significant RF effect (see Pitari et al., 2014). More important would be the effect on methane and other GHGs (see Vioni et al., 2017 and Tilmes et al., 2018) because of changes in photolysis rates and transport. Furthermore, in terms of RF, also the effects of a decrease in UT ice particles, as showed in Kuebbeler et al. (2012) and recently in Vioni et al (2018a), that would produce a cooling effect by trapping less planetary radiation, is important.

As we write we refer to limitations "*with SO₂ injection as a method of producing a radiative forcing perturbation*" like stated in former line 17-18. These limitations include "*effects that reduce the efficiency of the RF produced by the sulfate aerosols*" as stated by reviewer 1 above. We complemented the listing with tropospheric ice clouds and methane and removed ozone depletion from the listing.

Point (4) has nothing to do with RF and possible effects of SG on ecosystem have been studied in more details after Kravitz et al. (2012) (such as in Xia et al., 2017), together with other changes (tropical storm etc.). But this is not the point of this article.

Point 4 has been removed.

P. 4, line 8: Vioni et al (2017, 2018a and 2018b) also used a sectional aerosol approach in their model and fully coupled microphysics and chemistry. It would be good to compare some of their results with yours since they also focus on RF changes, as I explain in point 2) and in some of these comments.

The three papers you mentioned are now discussed as well and the model which was used is described in one sentence.

P. 4, line 19: Mills et al. (2017) is more of a description of WACCM-CESM new model set-up and the validation with Pinatubo data, so it is not about SG scenarios.

Corrected.

P. 6, lines 11-13: Here the authors should give the reader some more context on this. The amount injected in this work is not really an issue here (because you are doing sensitivity studies and not looking at the long-term climatic response), but it's a bit of a stretch to say that that's how much Pinatubo injected. We don't know exactly how much SO₂ Pinatubo injected, so from a modeling perspective the amount of SO₂ for Pinatubo is very model dependent (from 10 to 20 Tg-SO₂ to get the best agreement with AOD observations), and it would be good to mention this (Timmreck et al. (2018) would be enough to reference, but just look at, for instance, Mills et al. (2016) where they inject 10 Tg-SO₂ and Pitari et al. (2016), where they inject 20 Tg-SO₂). Just give some context to the reader. Personally, 1.83 Mt-S per year seems very arbitrary, and I would have preferred an amount more comparable to previous simulations.

The initial idea was to emit 2 Mt/yr, but due to a problem in the emission scheme, effectively only 1.83 Mt/yr have been emitted. As you already mentioned, this has no effect on the conclusions made in this paper as the same amount of sulfur has been emitted in all our model simulations. The 1.83 Mt/yr correspond to about 9-20% of the amount of S Mt Pinatubo emitted during the eruption 1991 as there are many different estimates ranging from 10-20 Mt S, depending which model one considers.

P. 7, line 2: I would suggest explaining a bit what the authors mean by "surf zone" and offer some references (as you do, rightfully, for the BDC). It might not be such a common term as you think.

Was corrected to "...emitting partly into the stratospheric surf zone and not only into the tropical pipe. The stratospheric surf zone is the region outside the subtropical transport barrier where breaking of planetary waves leads to quasi horizontal mixing (McIntyre and Palmer, 1984; Polvani et al., 1995)."

P. 12, line 1: As per my point 1): most other studies (see Tilmes et al., 2017, but also the entire GeoMIP G4 experiment) inject at only one longitude. In other simulations the single longitude was found either better for the scope (r_{eff} closer to the desired one, Tilmes et al., 2017) or unimportant (because of the fast mixing time). You should mention this here.

We refer now to other studies which have previously shown that point injections result in larger aerosol burden, r_{eff} closer to the desired size and larger short-wave radiative forcing values, i.e. Niemeier et al. (2011) and Tilmes et al., (2017).

P. 12, line 5: The shared longwave surface anomaly doesn't really say anything, especially since you don't have a surface coupling. It would not be due to the aerosol absorption anyway, but it is the result of many more processes. I suggest removing this phrase or explain better what you think the surface LW anomaly tells you, if you think it's important.

The sentence about the long-wave anomaly has been removed.

P. 12, line 14: constant climatological what? Concentration? It's confusing.

Corrected to: "Kleinschmitt et al. (2018) applied a mean lifetime of 41 days for SO₂ to H₂SO₄ conversion in their study and found a SO₂-to-H₂SO₄ conversion rate of 96 %."

P. 12, line 20: split this in two phrases, one for AS and one for CS. Don't use parenthesis this way.

Corrected. I split the sentence into two.

P. 13, line 5: Either you write "Emission strategies like that investigated by ..." or "Emitting at ... as investigated by ..."

The sentence has been rewritten.

P. 13, line 16-23: Again: ozone and water vapor are only two of the SG side effects when it comes to RF: you should at least mention the fact that methane lifetime would increase and ice cloud decrease.

We now explicitly state that only the negative side effects investigated in this study are considered in Table 3. Additionally, we looked at changes in methane lifetime, and point to other side effects (i.e. cloud feedbacks) which could have an inhibiting effect on solar geoengineering efficiency but are not considered here.

P. 14, line 1: like in the case of SO₂ emissions

Corrected.

P. 15, lines 7-8: as I said in my major point 1), mention the big difference between the studies, that is the different longitudinal distribution of the injections.

We now compare our point emission scenarios to Niemeier et al. (2011) and Kleinschmitt et al. (2018) and point out the difference between the studies.

P. 15, lines 13-14: Yes, but in the framework of CCMI, so considering reference scenarios. The response to the BDC to the stratospheric heating produced by SG is not something that is considered there. The reference is still useful but mention this, at least.

Corrected. I have added a sentence for clarification.

P. 15, line 24: As has been previously studied in Kuebbeler et al (2012) and Visioni et al (2018a).

References have been added.

P. 16, line 6: "with" current GCMs.

Corrected

Figure 2: This figure has a very poor resolution (compared to Fig. 5 that is similar). Panels b) and c) have such large scales and the curves are minuscule.

The resolution of all figures has been increased and the scale of panels b) and c) of figure 2 and panel b) of figure 5 has been adapted.

Figure 4: This is an interesting figure but must be improved. The scale for the SW forcing could be reduced a lot to highlight the differences between the different simulations (panel b from -1 to .3 W/m² and panel c from 0 to -2 W/m²). Furthermore, symbols are very hard to read. I suggest enlarging them (or using more color). Very minor thing, the is and ls in figure 4 are all "weird", they look bold while the rest of the letters don't. You should fix this. (same goes for figure 2, 6 and 7). It's probably just a problem of how you saved the figures.

The scale was adapted, and symbols were enlarged. The weird looking letters were replaced as well.

References:

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