Response to Reviewer # 1

Reviewer's comments are in blue. Author responses are in black.

General comment.

In the introduction, the authors already say that this question has been addressed by Kravitz et al. (2009), but they do not make a case for doing the same study again. What are the scientific questions that they are addressing that have not been answered before? Without this, I wonder why readers would want to wade their way through all the details in this paper of repeating the same experiment with two more models. Is there something about the current two models that would produce a more accurate simulation and have the potential to get a different result than before? If not, why do it? Yes, these models have an explicit nudged QBO, but would that be expected to produce anything fundamental that is different about sulfate deposition? Therefore, I recommend this paper be rejected as it has no new scientific conclusions.

We believe the reviewer is wrong in its initial judgement, starting from the title the reviewer gives to its review: "nothing new here". It is a matter of fact that any scientific study, starting from a pioneering one, tries to go further by analyzing important issues that were not considered in the first place. Otherwise, in the case of sulfate geoengineering, we would still be at the first pioneering study of Crutzen (2006). Or, if we only consider the issue of sulfur deposition, we could simply say that at the steady state what is globally deposited at the surface is what we inject in the stratosphere! Our study, on the other hand, is in our opinion a robust step forward with respect to the one of Kravitz et al. (2009), because it tries to better understand how stratospheric large scale transport and its periodic oscillations (i.e., QBO) may impact the cross tropopause sulfur fluxes and finally the regional sulfur deposition. This is what we call (even from the manuscript title) "quantification" of sulfur deposition.
We discuss what are the various complex mechanisms (dynamics and transport plus aerosol microphysics) driving the internannual changes of lower stratospheric and cross tropopause sulfur transport. The reviewer asks if the fact that the models have an explicit nudged QBO would "be expected to produce anything fundamental that is different about sulfate deposition": the results of the presence of the QBO are discussed in depth throughout section 3 and section 4.1 and 4.2 and in Figures 5,6,7,8 and 12. The connection between the QBO and the sulfate deposition is in particular detailed in Fig. 8 and 12, and in section 4.2. Only section 4.3 (Continental scale deposition) is dedicated to quantifying the deposition at a regional scale in a way that resembles the work of Kravitz et al. (2009), and we fully aknowledge it from the beginning. Furthermore, in the case of the aforementioned paper, they explicitly say that "that all the sulfur deposition is sulfuric acid" in their model, in order to calculate the upper limit of the sulfate deposition. In our simulations (see Table 4) we also tried to give an estimate for the share of SO_2/SO_4 and wet/dry deposition. Another point where our study differs from (and somewhat improves) the results from Kravitz et al. (2009) is the fact that their simulations were done with a prescribed effective radius. In our case, for ULAQ-CCM at least, we use a calculate size distribution for our aerosols and we analyze how the size of the sulfate aerosol population responds to stratospheric changes, and how this affects the optical depth (Fig. 7) and, lastly, the radiative forcing efficiency.
In conclusion, before responding to the single points raised by the reviewer, we disagree with the statement of the reviewer towards the rejection of this paper, on grounds that it is a repetition of an already published paper. We believe, indeed, that connecting changes in stratospheric

dynamics, sulfate aerosol microphysics and deposition is definitely a robust step forward, and that this is highlighted in multiple parts in our manuscript, in particular:

1) in Fig. 4 where we lay out the basis for our "advancement".
2) In Fig. 5 and 6 where we show how the tropical sulfate burden (and thus, as we show in Fig. 7, the AOD) is modified by the stratospheric dynamics. In Section 3, we conclude by looking at how these modulations are connected to different RFs. We also point out, in Section 3 and in the conclusions, that having an externally nudged QBO allows us to separate the feedback of the QBO in the confinement of the aerosols in a way that other studies (i.e. Niemeier and Timmreck, 2015), which have an internally generated QBO cannot, and how this might be important to better understand the underlying physical feedbacks.
3) In Fig. 12 where the microphysical and dynamical changes are highlighted by looking at the differences in strat-trop exchange during the different QBO phases.
4) Lastly, in fig. 15, where the regional deposition results are shown with the variability due to the discussed stratospheric changes.

Following this introduction, we will answer each issue the reviewer has raised, in hope that they (and any reader that might be interested) might be convinced of the goodness of our effort.

The abstract and conclusions use the metric of % of current sulfur deposition, but this relative value is not nearly as important as the total mass. Does the increased deposition in pristine regions represent a threat in terms of acid deposition on land or in the ocean?

The calculations are done throughout the paper in terms of total mass (Tg-S/yr; global, hemispheric, regional) (see Figures from 9 to 15 and Tables 2,3,4 and 5). In addition we show relative changes (in percent), simply because the community would like to know how much a potential implementation of a sulfate geoengineering SRM technique will affect local sulfur deposition values, relatively to the unperturbed background value. The reviewer suggestion to include also in the abstract the total mass metric will be taken into account by presenting there both absolute and relative calculated sulfur deposition changes. In the revised manuscript we shall also try to improve our final discussion towards deposition changes in pristine areas.

The global average results they get can already be easily calculated because in equilibrium 8 Tg SO2 into the stratosphere per year will produce the same surface deposition.

This seems rather obvious. At the equilibrium (and if the mass is conserved, as should be in a climate model) we have to deposit 8 Tg-SO_2/yr, if this amount is actually injected into the stratosphere. However, we show throughout the paper how significant interannual variations are produced by the QBO modulation of the stratospheric circulation, with induced changes in cross tropopause sulfur fluxes (in particular see Fig. 12, which shows the E-W phase average differences, produced by a full coupling of aerosol microphysics and large scale transport). This is indeed a robust step forward of our study, contrary to the reviewer general statements (see above).

p. 3, lines 31-33: The authors say they did a G4 experiment, but G4 required 5 Tg SO2 per year and not 8 Tg/year. This has to be corrected here and throughout the paper. G4 also had a 50-year emission and then a halt to emissions. The authors also have to explain why they chose to use 8 Tg/year.

Although the original definition of G4 in the Kravitz et al. (2011) paper is the one correctly reported by the reviewer, the overall meaning of the G4 experiment is to impose a fixed SO_2 injection, contrary to G3 where a time increasing injection is adopted in order to balance the increasing TOA positive radiative flux (essentially due to GHGs). We have adopted a "minor" adjustment of the original G4 definition by simply using a 8 Tg-SO_2/yr injection instead of 5 Tg-SO_2/yr and for a time period shorter than 50 years (even though the complete G4 numerical experiments run with the ULAQ model were carried out for 50 years plus 20 years termination period, as documented in Visioni et al., 2017b). The reviewer is right in asking us to explain why we chose a 8 Tg/yr injection: we did it in order to have, in ULAQ-CCM, the appropriate surface temperature taken from a fully coupled simulation run with the CCSM-CAM4 model that used a 8 Tg-SO_2/yr injection. In note 3 of Table 1 we briefly cite the aforementioned Visioni et al. (2017b) paper where we discussed this in depth. The choice of injection for GEOS-Chem follows in order to have a comparable injection to ULAQ-CCM. In the revised manuscript we will try to make it clear once again. In the conclusions, however, we already acknowledge (line 26-28, p. 34) that the proposed G4 injection by Kravitz et al. (2011) is 2.5 Tg-S/yr.

Why was the ULAQ model run with such low horizontal resolution but high vertical resolution? Does this affect the results? For example, how well is tropospheric deposition really simulated, as I would think the precipitation would not be expected to be able to address issues like rainout and washout of sulfate aerosols, and distinguish between wet and dry deposition? How well does it do this for the current climate with not geoengineering? Geos-Chem also seems to combine low horizontal resolution with high vertical resolution. Why?

Both ULAQ-CCM and GEOS-Chem use (in this study) a horizontal resolution close to T21, which we would not define as a "such low horizontal resolution". This is a fully acceptable resolution for studies focusing on stratospheric dynamics and transport and strat-trop exchange. Of course, it is possible to use higher horizontal resolutions, but this is not a strictly physical requirement. Many model intercomparison campaigns prove this (see for example SPARC-CCMVal-2 or CCMI). The use of a high vertical resolution is necessary to properly catch the tropopause altitude, due to the different aerosol behavior above and below the tropopause altitude. We do not understand the reviewer argument on wet and dry deposition. Both models account for wet, dry and gravitational deposition. Washout takes into account the contribution of large-scale and convective precipitation (see descriptions at page 9 lines 11-15 for ULAQ-CCM and page 10 lines 11-14 for GEOS-Chem).
Furthermore, regarding the reviewer question "How well does it do this for the current climate with not geoengineering?", we believe this is deeply explored in the manuscript considering we use Fig. 13 to compare our regional deposition results in the non-geoengineered case with two previously published results (Lamarque et al., 2013 and Vet et al., 2014) that used both multi-model ensembles and observations and Table 4 to compare our breakdown of wet and dry deposition to the same papers.

p. 9, line 22, starts in the middle of a sentence. Something is missing. How do you explain the longitudinal patterns of deposition changes in Fig. 11? Why are the depositions in the Northern Hemisphere much larger in the already polluted regions? Kravitz et al. (2009) also found this (their Fig. 2), at least for North America and Asia. Is this the region of maximum STE along tropopause folds and storminess, and just over the polluted regions by chance?

We apologize for the mistake at p.9, line 22. Yes, the words “The ULAQ-CCM” are missing, so that the correct phrase is “The ULAQ-CCM ability in producing a correct confinement of sulfate aerosols in the tropical stratosphere in SG or post-volcanic conditions has already been extensively tested…”

Regarding the deposition changes, we believe we have discussed this in p. 24, lines 10-13: “Non-zonal asymmetries of mid-latitude deposition flux changes result essentially from planetary wave modulation of the strat-trop downward flux, coupled to the precipitation frequency in the lower troposphere (see discussion below)” and lines 25-31: “Its maxima resemble a planetary wavenumber 1-2 modulation of the lower stratospheric poleward sulfate transport from the tropical pipe reservoir, thus consequently producing non-zonal asymmetries in the tropospheric sulfate influx. The tropospheric convective vertical mixing coupled to wet scavenging produces a tropospheric sulfate lifetime of approximately 5 days in the ULAQ-CCM (Pitari et al., 2016a). In a first approximation, zonal transport operated by the westerlies tends to move the downward moving sulfate coming from the tropopause by approximately 6500 km in a time period comparable to the tropospheric sulfate lifetime. This seems roughly consistent with the westerly displacement of mid-latitude sulfur deposition flux changes of Fig. 11a with the strat-trop sulfur downward fluxes of Fig. 12a.”

While it is true that some of those changes happen in already polluted zones, this is true for the East coast of North America and for the East coast of Asia also in our models, but not, for instance, Europe. This is because the reason is a combination of the position of the cross-tropopause downwelling with rainout at certain locations. This is in good agreement with the findings in Kravitz et al. (2009), as the reviewer states. We will be sure to add this in the discussion in the revised manuscript.

The authors would also need to address the 14 comments in the attached annotated manuscript.

1) P. 1, line 2: In the IPCC report from the Working Group III (Mitigation), Geoengineering is listed in Table 2.1 as a “other mitigation” scenario type, so we thought it was appropriate. However, following the reviewer advice, we will replace it with the word “offsetting”.

2) P.1, line 7-8: we agree, and will consistently add in the abstract also the absolute mass changes.

3) P.1, line 21: corrected.

4) P.1, line 23: we acknowledge this is imprecise and will correct it.

5) P.2, line 2: corrected.

6) P.2, line 5: we are aware of that. The error arose because the year refers to the republishing year of the chapter “The Climate of the Future” by the AGU (http://onlinelibrary.wiley.com/doi/10.1002/9781118665251.ch7/summary) but the year should definitely be 1977 which is when it was published for the first time.

7) P.3, line 10: changed.

8) Table 1: “pressure”, corrected.

9) Table 1: “Online” instead of nudging: considering that GEOS-Chem is a CTM, and that it is driven by assimilated meteorological fields (in this case, MERRA reanalysis), we believe it is not correct to say that there is a “nudged” QBO, that for ULAQ-CCM means that the latter model calculated its own circulation (being a CCM), but then is nudged towards a certain QBO. So we believe that the use of two different words is correct. We will further expand on this in the appropriate sections (2.1 and 2.2) to make it clearer.

10) Page 10, line 22: “The ULAQ-CCM” added, as discussed above.

11) Page 33, line 3-4: as we explain right after, we meant to say that the cooling effect of injecting sulfate in the stratosphere, given what it has been measured in case of explosive volcanic eruptions, would be a rather certain effect. However, we agree that a rephrasing would clarify our meaning and make it more correct.

12) Page 33, line 5: corrected.

13) Page 34, line 10-11: We are not sure about what the reviewer means: As we explain in the manuscript (see for instance table 2), our emissions consider both anthropogenic and natural (DMS) emissions, so that the global Base deposition is a result of both human and natural emissions. Therefore, our changes in case of SG are calculated against the sum of those two. If it was not the case, Fig. 13 (where we compare our Base results against Lamarque et al., 2013 and Vet et al., 2014, where they consider both natural and human emissions) would not give correct results.

14) Page 36, line 13: corrected, as discussed above.