Overview: This study discusses a new potential approach for sulfate geoengineering using an enhancement of COS at the surface. The study is very well organized, clearly written, and nicely presented. The paper should be published as an important contribution to possible approaches for sulfate geoengineering and their effects. I am supporting the publication after the authors considered the following points listed below. While this is a minor revision in terms of workload, I am suggesting major additions to the discussion and the abstract.

Here are some major concerns considering this new approach that has not been clearly addressed in the paper:

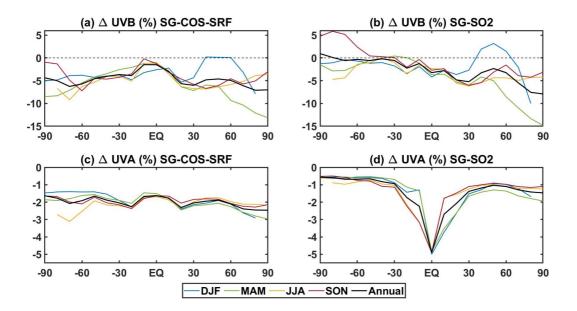
1) COS is more uniformly distributed in the troposphere and has a much longer lifetime than sulfate. This however suggests that there is much less control using COS than for example using SO2 injections for sulfate geoengineering and limits the potential use of a feedback control algorithm to modulate the amount of cooling in different hemispheres. However, earlier studies by Kravitz et al. (2017) have shown the (game-changing) potential of using a feedback control algorithm to reduce side-effects for instance to reach surface temperature targets or other impact-relevant targets. This is a major drawback of this approach in addition to the lack of the rapid regulation of the injections in case of a large volcanic eruption as already pointed out in this paper.

We have expanded the final discussion to better clarify this point raised by the reviewer.

2) Due to its toxicity, there is seems to be a hard limit in using COS. This study increased surface COS concentrations to reach 35ppb, which is very close to the possible limits of 50ppb, as stated in the text. The first concern that needs to be at least mentioned for future work is to identify how reliable the studies are that estimate this toxicity considering potential long-term increases of COS. Secondly, the enhancement of COS reduces AOD by 0.08 with a radiative forcing of -1.3W/m<sup>2</sup>, which may be translated to less than 0.5 degrees of cooling. Due to the uncertainty in different models, the cooling could be less and may not be sufficient. Furthermore, due to the long phase-in time, it will take a long time to find out how much cooling can be achieved. What if more cooling is needed? Would COS be eventually replaced by SO2 injections? What is the point then of using it in the first place, while adding the danger of exposing humans and ecosystems to a toxic pollutant? To me, this is a major issue that may be a showstopper for considering this method.

Given the feedback received, we have considerably expanded the discussion in places to further consider the problems that would be encountered with this approach, highlighting uncertainties in the response. Our new set of experiment, which consider the injection of COS below the tropopause (6 Tg-S of COS at 16 km of altitude and at the equator (0°N)), further highlights the possibility of using COS instead of other precursors.

3) It needs to be more clearly stated that surface UV is largely reduced with this method, and this can be harmful to humans and ecosystems. A more detailed discussion with the region and season would be helpful.



**Fig. 8**. Zonal UVB and UVA surface changes per each season in percentage with respect to BG case in SG-COS-SRF (panels a and b, respectively) and SG-SO2 (panels c and d). All quantities are averaged over the years 2046-2055.

We have updated Fig. 8 (see above) and the related discussion to further consider seasonal changes and to highlight the impact of UV changes at the surface.

**Detailed comments:** 

Abstract: The last sentence in the abstract stated that COS emissions are feasible. It may be technically feasible, but I think, the authors need to also point to the drawbacks of this approach in the abstract and conclusions, including the limitations compared to stratospheric SO2 injections.

We have updated the manuscript, also in light of the new set of experiments, to better clarify challenges and limitations with this approach.

Line 19: I don't think, this type of intervention can be classified as a "short-term" intervention, earlier studies have shown, that even a "Peakshaving" scenario may require injections between 80-160 years (Tilmes et al., 2016, 2020).

We removed the term "short-term".

Line 22: "optically active" is somewhat strange. You could maybe say, "the aerosol layer is thickened and therefore reflects more sunlight..."

We have substituted with "to obtain a cloud of aerosols capable of reflecting a portion of the incoming sunlight"

Line 27: I don't understand what you mean with "any proposed compound would quickly react to form sulfate aerosols", only sulfur will form sulfate aerosols. Other components may be coded with sulfates that are in the atmosphere. Is that what you mean?

Any compounds referred to the sulfur species that could be injected, including the largely discussed  $SO_2$  injection and the injection of  $H_2SO_4$  vapor (Pierce et al., 2010). COS falls in the same category.

Line 38: Is this the tropospheric lifetime? What is the stratospheric lifetime.

35 years is the tropospheric lifetime which includes only photochemical reactions (no land sinks). The stratospheric lifetime is about 10 years (due to an increase in photolysis). If land sinks are included, the overall lifetime is 3.8 years. We tried to clarify this in the manuscript.

Line 65: Since you are looking at UV, please also state what photolysis scheme is used in the model and if photolysis varies with aerosol concentrations.

As we detail in Section 4, we used the most recent version of the TUV code to determine UV changes at the surface.

Line 72: Is there a reason why increase COS emissions are placed at the same locations as the anthropogenic emission?

We placed the increased COS emissions according to the distribution of industrial sources of  $CO_2$  and  $CS_2$ , as those would be the likely sources used to eventually produce the larger fluxes of COS.

Line 101: Could you add an estimate of how much surface cooling one expects with an increase in 0.08 AOD? I would be probably less than half a degree of cooling considering the related GeoMIP experiments, is that correct?

We added a reference to Visioni et al. (2021), where such an evaluation was performed for G6 models. We added the following phrase: "*In the GeoMIP G6sulfur experiment* (*Visioni et al., 2021b*), the average global surface cooling reported by 6 Earth system models for a similar stratospheric OD was 0.46 K."

Line 104: Didn't you state that the lifetime is 35 years, not 3.8 years? What is different here?

The purely tropospheric **chemical** lifetime of COS is 35 years and decreases to 3.8 years when including both chemical reactions and dry deposition.

Line 106-107. I don't follow, what is meant here. 0.8ppbv of what, and why do you refer to the RF of other greenhouse gases? Do you mean, that after 20years, COS values have declined close enough to the background to not have a significant impact on the RF?

0.8 ppbv refers to the tropospheric concentration of COS. We want to emphasize the role as a greenhouse gas in the decreasing phase when sulfate aerosols are still in the stratosphere reflecting solar radiation. By comparing the radiative forcing of COS with that of other greenhouse gases that we have included in the RF discussion, we want to say that COS does not produce a significant impact on RF.

We changed "This means an increase of 0.8 ppbv with respect to background condition, that would produce a direct RF negligible if compared to other well mixed greenhouse gases." to:

"This means an increase of COS of 0.8 ppbv with respect to background condition, that would produce a direct RF negligible."

Line 230: The catalytic NOx cycle is decreased with more surface area density, which results in less ozone loss. Isn't the inhibition of denitrification more important in high latitudes and for cold temperatures, and less in other latitudes

As shown in Tilmes et al. (2017), NOx cycle changes are far more important at low latitudes but we've tried to clarify this in the revised manuscript.

Line 234: "its photodissociation" what is "its" referred to here?

"Its photodissociation" refers to the ozone. We changed the sentence to make it clearer like this:

*"Increasing stratospheric ozone affects UVB at the surface because it is absorbed by ozone during its photodissociation."* 

Line 240: change "UVA decrease is everywhere negative in both SG experiments" to "UVA decreases everywhere in both SG experiments"

## Corrected.

Table5 and Figure 8: You are nicely showing total column ozone variations with region and seasons, and it is very clear that there are differences in sign in the response. Illustrating UV changes annually is not a very meaningful measure especially for high latitudes. I would strongly recommend expanding this figure and showing 4 seasons instead or in addition to the annual values. I would also expand the discussion on the UV impacts, and why UV-A and UV-B and shown separately. Not sure if there is any reference to Figure 8 in the text?

This is discussed in conjunction with item (3). We have amended the text accordingly.

Line 269: reference should be Bruhl et al. (2012)

Apologies. We have fixed the name in the paper and references.

Line 289: should be (Figures 9 and S6)

Corrected.

## References

Pierce, J. R., Weisenstein, D. K., Heckendorn, P., Peter, T., and Keith, D. W. (2010), Efficient formation of stratospheric aerosol for climate engineering by emission of condensible vapor from aircraft, Geophys. Res. Lett., 37, L18805, doi:10.1029/2010GL043975.