Reviewers' comments are in bold. Authors' responses are in blue.

Overview: This paper proposes using carbonyl sulfide (COS) released at the surface for increasing stratospheric sulfate aerosol. This proposal has the advantage of not requiring distribution by stratospheric aircraft. The paper gives a great amount of detail on the differences between the three scenarios examined, a background, a geoengineering scenario using SO2 and one using COS. It is generally well written. I recommend publication after addressing the comments below, and most importantly, comment #1.

We thank the reviewer for their supportive comments. We have responded below to each point.

1) Crutzen (2006) https://link.springer.com/article/10.1007/s10584-006-9101-y states "An alternative may be to release a S-containing gas at the earth's surface, or better from balloons, in the tropical stratosphere. A gas one might think of is COS, which may be the main source of the stratospheric sulfate layer during low activity volcanic periods (Crutzen, 1976), although this is debated (Chin and Davis, 1993). However, about 75% of the COS emitted will be taken up by plants, with unknown long-term ecological consequences, 22% is removed by reaction with OH, mostly in the troposphere, and only 5% reaches the stratosphere to produce SO2 and sulfate particles (Chin and Davis, 1993). Consequently, releasing COS at the ground is not recommended."

Based on this, first, this paper should note that this has been proposed before (and therefore may not be so novel) , and discounted due to potential ecological damage. There is discussion at the end of the paper that this sort of thing should be looked at. That discussion should be at the start of the paper.

We expanded the discussion about ecological impacts following the suggestions of point 1) and we added concerns in the introduction about the unknown effects on the efficiency of the uptake by plants and soils in case of higher concentrations of COS, as also suggested by the comment of Dr. Whelan. We added the following phrases:

"Not much is known however about the response of ecosystems in the presence of high concentrations of COS: Stimler et al. (2010) showed that high levels of COS enhance the stomatal conductance of some plants, which might in turn have other unforeseen effects; further, Conrad and Meuser (2000) proposed that high COS concentrations may interact with soils and possibly change soil pH"

Further, following comment 6) from the reviewer, we also analyzed a new set of simulations with injections of COS right below the tropopause, that might allow to bypass part of the concerns regarding too high surface COS concentrations. The manuscript has been modified in multiple places to include the new results.

Second, is the balance between global warming due to COS vs cooling due to aerosols produced taken into account in this study? Bruhl et al. 2012 state

"Further, using a chemical radiative convective model and recent spectra, we compute that the direct radiative forcing efficiency by 1 kg of COS is 724 times that of 1 kg CO2. Considering an anthropogenic fraction of 30 % (derived from ice core data), this translates into an overall direct radiative forcing by COS of 0.003 W m−2 . The direct global warming potentials of COS over time horizons of 20 and 100 yr are GWP(20 yr) = 97 and GWP(100 yr) = 27, respectively (by mass). Furthermore, stratospheric aerosol particles produced by the photolysis of COS (chemical feedback) contribute to a negative direct solar radiative forcing, which in the CCM amounts to −0.007 W m−2 at the top of the atmosphere for the anthropogenic fraction, more than two times the direct warming forcing of COS. Considering that the lifetime of COS is twice that of stratospheric aerosols the warming and cooling tendencies approximately cancel." They also say "Therefore, if we account for indirect chemical effects in GWP calculations, also customary for gases such as methane (IPCC, 2007), it follows that COS has almost no net climate impact." So, if the warming and cooling tendencies cancel, is there an actual advantage to increasing COS emissions? At line 280 this paper states that the COS case produces an RF of .17 W/m2. What is the RF due to the increased sulfate aerosol produced by the COS? How do these results compare to the conclusions of Bruhl et al. 2012?

In the revised version, we have tried to clarify this point.

We estimate the RF due to the enhanced COS based on the definition of GWP as in Bruhl et al. (2012) and the result is a global warming uniformly distributed latitudinally of 0.17 W/m^2.

Under clear sky conditions, we calculated a net clear sky forcing of -2.01 W/m^2 due to scattering of solar radiation by sulfate particles (and absorption of LW). This is almost uniformly distributed latitudinally (fig. S6 in the original supplementary) as it follows the distribution of the aerosols and their effective radius (r_eff). The value of stratospheric r_eff determines the efficiency of the interaction of sulfate aerosols with the solar radiation, which peaks at a value of 0.5 μm. In SG-COS the stratospheric r_eff has a global value of 0.46 μm (compared to 0.1 to 0.2 μm in the unperturbed atmosphere in Bruhl et al. 2012 and to 0.52 in SG-SO2) and it is uniformly distributed at all latitudes.

When the contribution of background clouds and cirrus thinning is included, the overall net forcing is -1.52 W/m^2 that is more than 2 times greater than the direct warming forcing of COS in the background condition in Bruhl et al. 2012.

We have revised the manuscript to say: "*Overall, this results in a radiative forcing from the COS increase of 0.17 W/m^2.*

The main contributions of sulfate aerosols and clouds are summarised in tables S5 and S6 for SG-COS and SG-SO2, respectively. The contribution of sulfate aerosol is the sum of the cooling effects given by the efficient scattering of solar radiation by particles of radius of 0.5 μm and the absorption of LW by larger ones.

Globally, the estimated values are similar for the Clear-Sky SW and LW forcing from the sulfate aerosols: in terms of the latitudinal distribution, however, SG-SO2 presents a peak in the tropics whereas the forcing from SG-COS is much more latitudinally even.

The reduction in optical depth from cirrus clouds (see table 1) produced by the aerosols (Kuebbeler et al., 2012; Visioni et al., 2018a) results in a net negative radiative forcing.

This is given by the balance between the positive RF in the shortwave (SW) due to the reduction of reflected solar radiation and the negative RF in the longwave (LW) due to the decrease of trapped planetary radiation, which reduces the contribution to the greenhouse effect. "

2) The text makes the statement "it is not a toxic gas for humans or ecosystems: negative effects have been found only when concentration exceeds 50 ppm (100,000 times more than the background mixing ratio (Kilburn and Warshaw, 1995; Bartholomaeus and Haritos, 2006)." The Bartholomaeus and Haritos paper does not have that statement, and the Kilburn and Warshaw paper is about H2S and not COS. Different references are needed to say that there are no negative effects on plants and ecosystems.

The reviewer is correct on both counts. In the revised version, firstly we have removed the mention of ecosystems and focused on human health. For the latter, we have removed the reference to Kilburn and Warshaw, and added the reference to Svoronos and Bruno, 2002, which is where the value of 50 ppm was from (and that is later cited by Bartholomaeus and Haritos, 2006). In the 2002 work, the authors state "Chronic exposure of 50 ppm (mass/mass) carbonyl sulfide (to noncholesterol fed rabbits) of between 0.5 and 12 weeks showed no histotoxic effect on the intimal or subintimal morphology of coronary arteries or the aorta. Similar studies of 50 ppm (mass/mass) carbonyl sulfide exposure to rabbits for 7 weeks showed no effect on the myocardial ultrastructure, and only a slight elevation of the mean serum or aortic cholesterol concentration was observed."

We have revised the manuscript to say: "*In the concentrations found in the atmosphere, it is not a toxic gas for humans: negative effects have not been found even at around 50 ppm, which is 100,000 times more than the background mixing ratio, and for long exposure times in mice and rabbits (Svoronos and Bruno, 2002). Higher concentrations than that can, however, be harmful (Bartholomaeus and Haritos, 2006)"*

3) The Budyko, 1978 reference link (Budyko, M. I.: The Climate of the Future, American Geophysical Union, https://doi.org/10.1002/9781118665251.ch7, 1978.) is broken (it says the doi cannot be found, this is apparently an error on the AGU web page.). I believe the publication date is actually 1977, and you can get to the full book at https://agupubs.onlinelibrary.wiley.com/doi/book/10.1029/SP010. The reference should be to the full book, since chapter 7 doesn't address the point being made. And, in support of the text (actually proposing SRM) NAS 1992 (National Academy of Sciences (NAS): 1992, Policy Implications of Greenhouse Warming: Mitigation, Adaptation, and the Science Base, Panel on Policy

Implications of Greenhouse Warming, Committee on Science, Engineering, and Public Policy, National Academy Press, Washington DC, 918 pp, https://www.nap.edu/catalog/1605/policy-implications-of-greenhouse-warming-mitigation-adaptation-and-the-science should be added.

We thank the reviewer for catching the broken reference. We have updated it as suggested and added the one to the 1992 NAS report.

4) Line 72: It should be noted at the beginning of the paper how the increase of 40 Tg-S/yr compares to current emissions. I finally found the factor of 400 in the conclusions.

We added the information regarding the emission of COS in background condition and the reference to the table in the Supplementary Material where the globally-annually averaged COS sources and sinks are listed for BG and SG-COS.

"The first geoengineering experiment, SG-COS, tries to produce a significant stratospheric aerosol burden by enhancing current anthropogenic surface emission sources of COS (0.12 Tg-S/yr, see table S1) by up to 40 Tg-S/yr."

5) Line 105-106 says "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." First, change to "a direct RF that is negligible compared to other well mixed greenhouse gases." Second, is the RF negligible to the negative forcing caused by the increased stratospheric aerosols?

We have changed the phrase as suggested. In 2075 there is still an increased OD from the stratospheric aerosols, so the direct RF from the GHG is still smaller than that from the aerosols. We have added this to the previous phrase.

6) And, another experiment that could be run. Instead of emitting a large amount of OCS at the surface, what about emitting at the tropical tropopause? It would require significantly less material, and may produce similar results, with a higher aerosol layer and similar latitudinal distribution. It would also avoid any issues with ecological damage due to increasing surface COS amounts.

We thank the reviewer for this suggestion. We ran a new set of experiments injecting 6 Tg-S of COS at 16 km of altitude and at the equator (0ºN) to test such an hypothesis. We found that the same stratospheric burden of sulfate can be achieved with substantially less injection of COS (and much less increase in tropospheric COS). The manuscript has been substantially changed in places to reflect the new results.