Response to Reviewers' Comments

Injection Near the Stratopause Minimizes the Stratospheric Side Effects of Sulfur-Based Climate Intervention

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We thank very much the reviewers for their helpful comments. The response to each reviewer's comment is marked in blue.

RC2

Review by Thomas Peter

General comments

This is an interesting manuscript on a new idea for how climate intervention through stratospheric aerosol injection (SAI) could be implemented by injecting at much higher altitudes (~50 km) than previously proposed. This could lead to significantly less harmful side effects than previous proposals to inject SO2 into the lower stratosphere. This could reduce both the strong warming of the lower stratosphere due to IR absorption by H2SO4-H2O aerosol, which affects climate and precipitation zones in the troposphere, and the depletion of stratospheric ozone. As the authors describe in their manuscript, this new idea could even boost the efficiency of surface cooling.

To my knowledge, this idea is original, and the proposed method could potentially be very important and would fit well into ACP. The authors are to be commended for developing this concept. Unfortunately, however, I do not believe that the technical details necessary to justify the feasibility of this novel method are sufficiently developed for publication in ACP.

Thanks Prof. Thomas Peter for the constructive suggestions, which help to improve the manuscript. Point-to-point responses are marked in blue.

The differences in aerosol formation at an altitude of 50 km compared to 25 km are considerable and would need to be discussed:

(1) The temperatures are so high that the formation of H2SO4-H2O droplets close to injection altitude seems unlikely.

(2) The air density is so low that the fall velocity of particles, if they form, will be very high, which is a major limitation.

(3) The H2SO4 photolysis, which is neglected in this modeling, could massively alter the model results.

These three aspects are not addressed in the submitted manuscript (with the exception of a reference to the fact that H2SO4 photolysis is neglected). However, the issues associated with these aspects are central to such a new proposal and must not be ignored. Therefore, I do not think that the manuscript can be accepted for publication in its present form.

Sorry for the confusion. We think all three aspects stem from one potential misunderstanding that needs clarification:

We inject SO_2 at 50 km, but our simulations show that sulfate aerosol formation occurs at much lower altitudes. Figure R3 shows the simulated zonal and vertical distributions of SO_2, sulfate aerosol, and H_2SO_4 anomalies in both SAI_{25} and SAI_{50} scenarios. In SAI_{25}, the simulated aerosol peak is located around 25 km in the tropics (the injection region). However, in SAI_{50}, the sulfate aerosol forms at lower altitudes, with peak concentrations below 20 km at high latitudes. The simulated H_2SO_4 anomalies are several orders of magnitude smaller than SO_2 at 50 km. We chose to inject SO_2 at 50 km because the overturning circulation rapidly transports these precursor gases poleward, leading to a more uniform global distribution. In the revised manuscript, we add Figure R3 into the supplement.

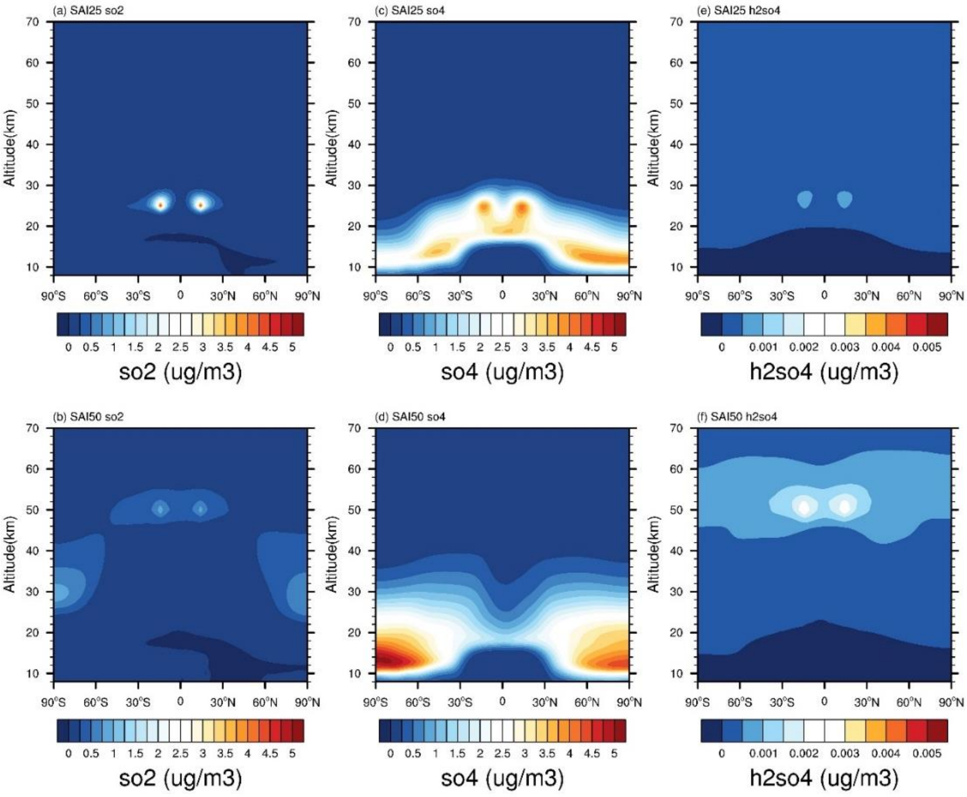

Figure R3: (a) The vertical distribution of the zonal and annual mean SO_2 anomalies in SAI_{25}. (b) same as (a) but for SAI_{50}; (c-d) same as (a-b) but for sulfate aerosol. (e-f) same as (a-b) but for H_2SO_4. Note that the contour range in panel (e-f) H_2SO_4 is 3 magnitudes smaller than sulfate and SO_2.

Detailed responses to each specific aspect are shown below in the Specific Comments section.

Specific comments

In the following, I will first explain my concerns regarding these three points in more detail and then provide a list of minor comments and suggestions for improvement of the manuscript.

Regarding (1): At the stratopause, temperatures range between 260 and 270 K. At such high temperatures, the normal Junge aerosol can no longer exist. The H2SO4 vapor pressure of aqueous sulfuric acid is approximately 10-8 hPa (see Fig. 4 of Carslaw et al., Revs. Geophys., 35, 2, p. 125,1997). At the stratopause this would correspond to 10 ppbv H2SO4 in the gas phase, i.e. about 100 times the total mixing ratio normally present in the lower stratosphere during volcanically quiescent periods. I think the mixing ratio reached by SAI50 would remain lower. Therefore, the nucleation of sulfuric acid particles probably only begins well below the injection height. Figure 1C seems to confirm that. How does the model treat this nucleation? No information is provided about the microphysical code, which for this proposal should be of major concern. I suppose the microphysics is treated by a modal approach, and it would be important to see size distributions at various altitudes.

The reviewer is correct that the formation of sulfate aerosol is difficult at stratopause near 50 km. As shown in Fig.R2, the simulated sulfur-species at 50 km in SAI_{50} scenario is dominated by SO_2, rather than H_2SO_4 gas nor sulfate aerosols. The simulated H_2SO_4 gases is 3 orders of magnitude lower than SO_2 at 50 km in SAI_{50}. Aerosols form when SO_2 gases are transported to higher latitudes and lower altitudes, with peak aerosol concentrations simulated in lower-middle stratosphere (Fig.R3). We added Figure R3 to supplement file (Figure S3), and explained this point in Line 151:

"Note that the sulfate aerosol evaporates into sulfuric acid gas above 35-40 km but reforms when the gas is transported to lower altitudes (10-30 km) via large-scale circulation."

In Line 138-141 :

"…It's important to note that while SO_2 is injected at 50 km, the actual sulfate aerosol formation occurs at much lower altitudes (primarily between 10-30 km) due to the rapid transport of precursor gases and more favorable conditions for aerosol formation at lower altitudes. Above 40 km, the simulated stratospheric sulfur species primarily exist in the form of SO2, with ~3 orders of magnitudes higher than H2SO4 (Fig. S3). Above 40 km, the simulated stratospheric sulfur species primarily exist in the form of SO_2, with ~3 orders of magnitudes higher than H_2SO_4 (Fig. S3).”

We also describe aerosol scheme MAM3 in the method section in Line78-81:

“MAM3 provides a physically-based treatment of aerosol size, mixing, and key microphysical processes, including nucleation, growth, deposition, and interactions with clouds and precipitation (Liu et al., 2012). The nucleation of sulfate aerosol is produced from aqueous-phase SO_2 oxidation and to a lesser extent from H_2SO_4 condensation on pre-existing aerosol (Liu et al., 2012).”

Regarding (2): The air is so thin at the injection height that particles with a radius of 100 nm sediment by about 10 km within a month (eyeballed from Fig. 2 of Müller & Peter, Ber. Bunsenges. Phys. Chem. 96, p. 353, 1992). Since this is a fundamental aspect of the proposed injection scheme, it would be necessary to check the model's implementation of this process carefully and to provide arguments, why this fast sedimentation does not invalidate the whole procedure.

We agree that aerosol sedimentation is faster in thinner air at higher altitudes. However, in our SAI_{50} simulations, the injected aerosols do not remain near 50 km but accumulate around 25–30 km altitude (see Fig. 1c).

We also confirm that CESM-WACCM can represent gravitational settling, and this process has been validated against observational estimates of aerosol lifetimes from major volcanic eruptions, such as Pinatubo and Hunga Tonga (Figure 1a).

Regarding (3): The photolysis of H2SO4 molecules is a central process in this scheme and cannot be ignored without good reason. A quick test with our own chemistry-climate model shows that the amount of condensed H2SO4 in the Junge aerosol layer is 2-4 times higher without photolysis than with photolysis. I would estimate that the reduction of aerosol mass in the upper stratosphere could more than a factor of 10. I must therefore assume that in the modeling work shown, a significant portion of the aerosol is formed solely due to the lack of photolysis in the model.

Following the reviewer’s suggestion, we compared the vertical distributions of sulfur-containing species in our SAI simulations. As shown in Figure R3-4, the simulated concentrations of H_2SO_4 are 1-3 orders of magnitude lower than those of sulfate aerosols.

We add H_2SO_4 photolysis into the model and compare the vertical distributions of the sulfur species with and without photolysis. Shown in Fig. R3b, the simulated vertical distributions of sulfur-containing gases and aerosols remain largely unchanged between simulations with and without photolysis. These results indicate that H_2SO_4 photochemistry has a limited impact on the overall sulfur distribution in SAI_{50} mainly because sulfate aerosols form in much lower altitude instead of stratopause, where the precursors SO_2 are transported rapidly by the overturning circulation.

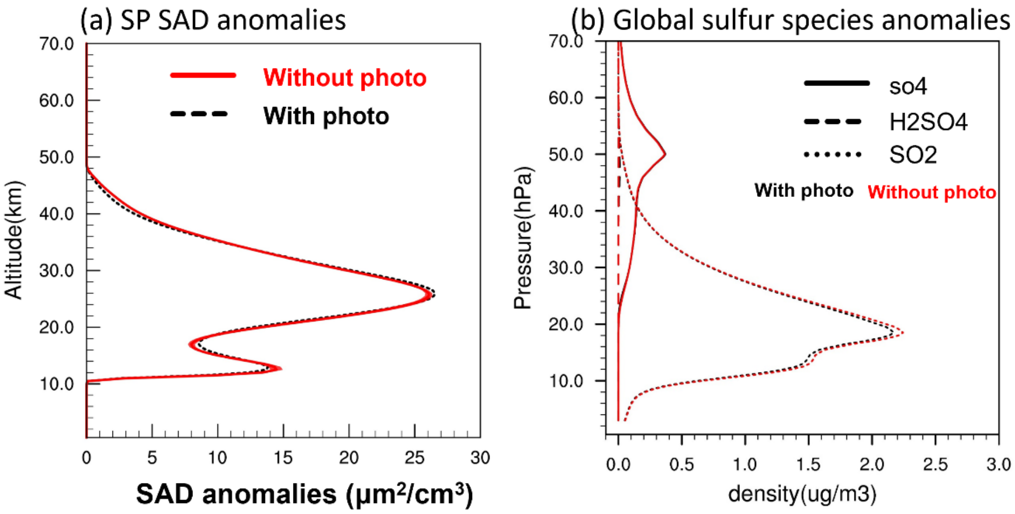

Figure R4. (a) Simulated vertical distributions of the Antarctic SAD with (black) and without (red) H_2SO_4 photolysis (same as Fig 1c red line) averaged from September-October-November (SON). (b) Simulated annual global mean vertical distribution of the sulfur species with (black) and without (red) H_2SO_4 photolysis. The simulated sulfate, H_2SO_4, and SO_2 are denoted by solid, dashed, and dotted line, respectively.

While points (1) and (2) can probably be positively resolved with the existing model runs (showing that the model correctly calculates the partial and vapor pressures of the aqueous sulfuric acid under upper stratospheric conditions and the sedimentation velocity of the aerosol particles formed), point (3) is likely to pose a bigger problem. If photolysis cannot be incorporated into the existing model, at least a very clear warning should be included for the reader that this omission may significantly influence the result and diminish the efficiency of the proposed method.

Please see the point-to-point response above.

Technical comments listed by line number

1. L. 19: “SAI using sulfur cools the planet” à “SAI using sulfur has been proposed to cool the planet”

Done

2. L, 20: Better don't talk about "traditional" SAI, in particular not in the first sentence of the abstract. All SAI is still in the proposal phase and largely unsubstantiated ideas, nothing traditional.

Revised to be "A commonly proposed SAI, with sulfur dioxide injection rate of 10 Tg/year at 25 km …."

3. L. 38: In addition to Ferraro et al. (2015) and Visioni et al. (2021), another excellent example making this point is Wunderlin et al., "Side effects of sulfur-based geoengineering due to absorptivity of sulfate aerosols", GRL, 2024.

   Done

4. L. 66: I do not understand the "positive ozone chemical tendency".

   We rewrite the sentence in Line 68: "By doing so, high-altitude sulfate aerosols reduce NOx levels, slowing NOx-driven ozone loss and allowing ozone to accumulate in the middle stratosphere, which can offset the ozone loss caused by reactive halogen species in the lower stratosphere."

5. L. 73: Here I expected more information on the type and characteristics of the microphysical module used in this modelling work

   We also describe aerosol scheme MAM3, in Lines 78-81:

   "MAM3 provides a physically-based treatment of aerosol size, mixing, and key microphysical processes, including nucleation, growth, deposition, and interactions with clouds and precipitation (Liu et al., 2012). The nucleation of sulfate aerosol is produced from aqueous-phase SO_2 oxidation and to a lesser extent from H_2SO_4 condensation on pre-existing aerosol (Liu et al., 2012)."

6. L. 79: I do not understand and do not accept that the fact that the column-integrated stratospheric burden of H2SO4 is much smaller that the burden of sulfate aerosols could be used for not having to take the photochemistry of H2SO4 into account. In the warm upper stratosphere, all H2SO4 is gaseous and exposed to H2SO4 + hv.

   As discussed in previous response, the simulated aerosol peaks in 20-25 km in altitude instead of 50 km. SO_2 is released at 50 km but transported polewards and downwards until sulfate aerosols are formed.

   We delete the sentence in the method section and address the H_2SO_4 photochemistry in the result section in Line 141-142:

   "…, and the simulated stratospheric sulfur species primarily exist in the form of sulfate and SO_2, with ~3 orders of magnitudes higher than H_2SO_4 (Fig. S3)

7. L. 84: “SO2 was continuously injected … with a total rate of 10 Tg per year”. It might be more meaningful to say “with a total rate of 27 Mg per year” to stress the continuous character, or even “with a total rate of 27 tons per year”.

   In the SAI experiments, SO_2 was continuously and uniformly injected throughout the year, with a total annual amount of 10 Tg injected per year.

8. L. 87: 5 years of model spin-up plus 15 years of actual model run. This is okay. But then, does Figure 1 show the 5 years of spin-up plus 10 years of model run?

   In the revised manuscript, Figure 1a is extended to 20 years, consistent with 15 years model run with 5 years spin-up.

9. L. 90: I do not understand these scaling factors. Where are they from?

   We rewrite in Line 99-101: “For simulations of year 2000, model is initialized with atmospheric ODS and Greenhouse Gases (GHGs) conditions of year 2000. For simulations of year 2040 (2065), the ODS and GHGs are fixed in the year of 2040 (2065).”

10. L. 103: The “2022 Hunga volcanic eruption”. Okay, everybody knows which volcano this is, yet it is a pretty crude abbreviate

    Revised to be “Hunga Tonga-Hunga Ha'apai”

11. L. 110: The “spread is designed to capture” sounds weird. Are you designing a spread or is the spread the result of your simulations?

    Thanks. Revised to be: “The spread across our simulations of 45 ensemble members represents the natural variations in stratospheric circulation.”

12. L. 127: Sentence confusing. For clarity, I would rewrite “… are similar for all lower altitude injections (at 20 km, 25 km and 35 km),...”.

    Done

13. L. 129: The word anomaly is used abundantly, also when it is just a “change” or even a total number. For instance, in Figure 1a is it really AOD anomaly or just AOD. And why is it in Figure 1C simply “SAD”, and not “SAD anomaly”?

    Thanks for the comment. Figure 1a, b, c are all AOD or SAD anomalies. We modified Fig.1c label to be “SAD anomaly”. We checked throughout the manuscript.

14. L. 260: “would be” instead “is”.

    Corrected

15. L. 295: Where is the dip in SAD (Fig. 1c) at 18 km come from?

We show the monthly vertical distribution of aerosols and found that the dip in SAD at ~18 km results from the interaction between transport and microphysical processes. Analysis of monthly vertical distributions reveals two distinct aerosol peaks:

- An upper peak (25-30 km) formed by continuous poleward transport of newly formed sulfate aerosols via the upper branch of the Brewer-Dobson Circulation (BDC)=
- A lower peak (~15 km) representing older aerosols that have descended through gravitational settling

During Antarctic spring (shown in Fig.1c), these two separate peaks create the observed dip at ~18 km. This pattern emerges as the BDC continuously brings new aerosols to high altitudes while previously transported aerosols settle to lower levels

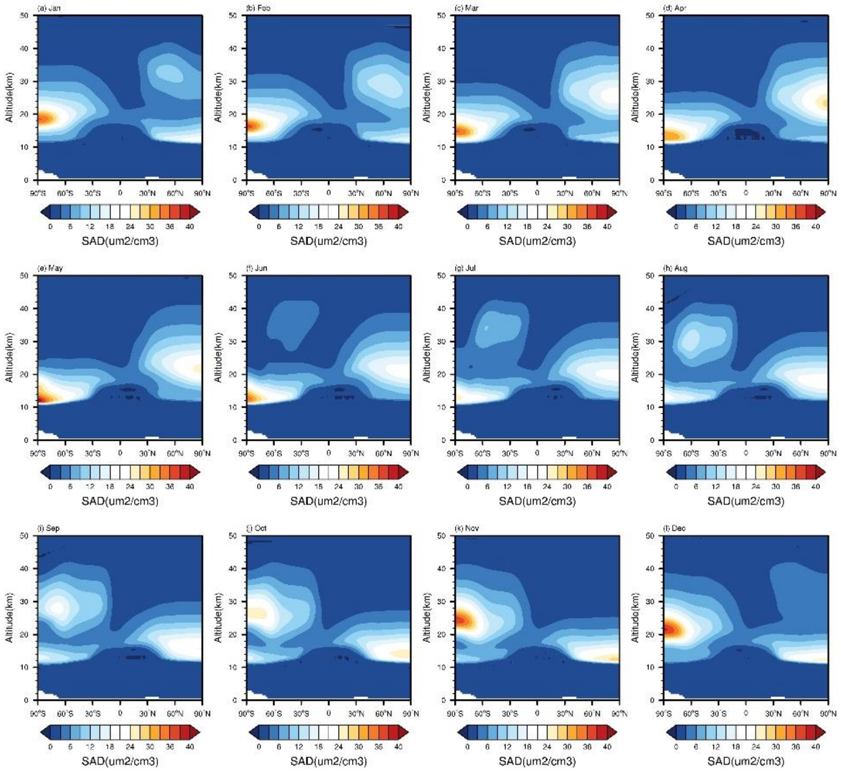

Figure R5. Simulated monthly vertical distribution of SAD anomalies in SAI_{50} from January (a) to December (l) after injection.

16. L. 295: The red curve in Fig. 1c would probably look quite different if H2SO4 + hv was taken into account.

We have conducted additional simulations that include H2SO4 photolysis (H2SO4+hv). The simulated Surface Area Density (SAD) vertical distributions are shown in Figure R3. While H2SO4 photolysis does affect the sulfate aerosol lifecycle, the overall vertical distribution pattern remains similar to our original simulations without photochemistry -

the simulated SAD still peaks between 20-30 km. This is because the primary formation of sulfate aerosol occurs at lower altitudes, and photolysis becomes more significant only at higher altitudes where the aerosol concentration is already low.

17. L. 296: Why distinct?

    Deleted

18. L. 301: “Multiple” is not a verb.

    Corrected

19. L. 305: “from ensembles” is slang. “of the ensemble members” would be appropriate.

    Corrected