

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 suggestions. The response to each reviewer's comment is marked in blue.

RC1

I'd like to thank the authors for considering and responding my comments. In many ways I think the manuscript has improved. I'm still not fully convinced of some of the replies.

I had remarked that the comparison of emissions at 50 km is only done with respect to equatorial emissions at lower altitudes while other emission strategies for the lower stratosphere exist. In response the authors have added three sentences to the introduction in which they refer to high-latitude emission strategies, and list some deficits of equatorial emissions that could be alleviated, among them "stratospheric warming". Now the main conclusion of this new manuscript seems to be indicated by the title: "Injection Near the Stratopause Mitigates the Stratospheric Side Effects ...". I think it is necessary to refer to alternative strategies not only in the introduction but also in the conclusions. How do the potential benefits of the stratopause emission strategies compare to other alternative strategies?

we added discussion in Line 275-278:

"Note that polar injection strategies also aim to mitigate tropical lower-stratospheric warming and preserve sea ice (Lee et al., 2021; Lee et al., 2023b), the SAI50 scenario requires less aerosol mass to achieve the same temperature target due to longer aerosol lifetime. In addition, the polar aerosol layer in SAI50 resides at higher altitudes than in polar injection scenario, which helps suppress NOx-catalyzed ozone loss and mitigate the severe ozone depletion caused by low-stratospheric aerosol accumulation."

I'm not convinced yet concerning the explanation for 22% greater cooling for 10% AOD increase in SAI50 compared to lower stratospheric injection. The authors now write this "primarily reflects the higher proportion of aerosols distributed at high

latitudes” and a “minor contribution also comes from the reduced stratospheric water vapor enrichment”. While I think this is possible, I don’t understand how this was analyzed and why the forcing dependence on surface temperature can be excluded as a contributor.

we rewrote in Line 187-195:

“In SAI50, the simulated 22% greater global mean surface cooling compared to the 10% increase in global mean AOD (Fig. 1a) reflects a combination of factors, including a higher proportion of aerosols distributed at high latitudes that enhances the efficiency of aerosol forcing through Arctic amplification processes, such as ice-albedo feedback and stable atmospheric conditions (Barnes and Polvani, 2015). In addition, reduced stratospheric water vapor enrichment (Fig. 2c–d) and the cooler Arctic surface (Hegde et al., 2025) can also contribute to the amplified Arctic cooling response.”

In their reply to my comment on implementation the authors argue their “study represents a theoretical exploration using idealized numerical experiments and focuses specifically on understanding the physical mechanisms and climate response …”. This seems inconsistent with several statements in the Summary and Outlook section. There the authors discuss how the injection could be done technically (“reusable rockets”, “benign emissions from a hydrogen fueled rocket platform”). So it seems to me the authors try to find arguments for their proposal and, by refusing to talk about costs, ignore potential arguments against it. I’m not talking about a detailed cost study, but if “the concept is well within the scope of current technology”, as the authors claim, it can’t be too hard to come up with a back of the envelope estimate.

we rewrote this part:

“Based on SSP2-4.5 scenario, achieving the 1.5-degree temperature goal would require an annual SO₂ injection rate of 3-8 Tg/year during 2040-2060 (Macmartin et al., 2022). Delivering 3-8 Tg of SO₂ per year to 50 km altitude could, in principle, be achieved with a fleet of 30-80 reusable rockets each with a 500-ton payload, and each launched every other day. Although detailed engineering analysis of a 50 km SAI injection suborbital launch system has not yet been done, the concept is technically plausible given current and emerging spaceflight technologies (Chang and Chern, 2021; Larson et al., 2017) and recent spaceflight experience. Indeed, the requirements of a SAI50 rocket-based injection system overlap with requirements and goals of other technologies such as rapid point-to-point rocket cargo that require low-cost routine operations (Chang and Chern, 2021). Our discussion is intended to highlight the potential plausibility and physical implications of high-altitude delivery, rather than to provide an engineering design or cost assessment, which would require dedicated analyses in future work.”

#RC2

The authors have largely addressed my concerns, but some points need further clarification.

In my first review, I had the following three main concerns:

- (1) From the original manuscript, I got the impression that the H₂SO₄-H₂O droplets proposed for SAI formed at the stratopause, even though temperatures at these altitudes are so high that the formation of H₂SO₄-H₂O droplets is unlikely.
- (2) Based on this impression, I criticized that the initial fall velocity of the particles would be very high, which poses a major complication for the model that would need to be addressed.
- (3) Finally, I believed that the photolysis of H₂SO₄, which was neglected in this modeling, could massively alter the model results.

In the revised version, the authors address these three concerns as well as all individual comments.

Concerns (1) and (2):

The authors clarify that my impression that the H₂SO₄-H₂O droplets in their model form at the stratopause was a misunderstanding.

The "Results" section now states: "It's important to note that while SO₂ 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." This is also clearly illustrated by the new Figure S3 in the supplementary information. Thank you. This is indeed in line with my expectations and does not change the potential value of the proposed new scheme. It also essentially addresses my concerns regarding points (1) and (2).

However, the false impression has not been completely dispelled. The fifth sentence of the abstract states: "In SAI50, the mean meridional overturning circulation near the stratopause rapidly transports aerosols to mid-high latitudes...". This still sounds as if the circulation near the stratopause transports aerosol particles to higher latitudes, which is not the case. Rather, the circulation near the stratopause transports gaseous SO₂ to higher latitudes, from where the SO₂ (plus some already formed gaseous H₂SO₄) is transported further to lower altitudes, finally oxidized completely to H₂SO₄ and forming H₂SO₄-H₂O droplets only below an altitude of 30-35 km (through bimolecular homogeneous nucleation or heterogeneous nucleation, e.g., on meteorite dust particles).

Thanks. It is revised to be:

"In SAI50, the mean meridional overturning circulation near the stratopause rapidly transports gaseous SO₂ to mid-high latitudes, preventing sulfate aerosol accumulation in the tropical lower stratosphere."

The confusion continues in the Introduction, with the following statements: "To minimize the Antarctic ozone loss, it is essential that some sulfate aerosols from the intervention remain at high altitudes in the polar stratosphere. By doing so, high-

altitude sulfate aerosols reduce NOx levels... In addition, aerosols formed at higher altitudes are rapidly transported to the mid-high latitudes rather than accumulating in the tropical lower stratosphere." I cannot see that aerosol particles, which form at higher altitudes, are then "rapidly transported to mid-high latitudes". I think the new Fig. S3d suggests instead that the particles are already at high latitudes when they nucleate and do not need to be transported there. This description has the potential to mislead readers and should be improved before publication.

We rewrote this sentence in Line73:

"In addition, sulfate aerosol concentrates in the mid-high latitudes rather than accumulating in the tropical lower stratosphere."

Concern (3):

In response to my concern that photolysis of H₂SO₄ must not be neglected as it could massively alter the results, the authors present simulations with and without H₂SO₄ photolysis. First, I am surprised that they can do this so easily, as I had assumed that the previous neglect was due to the model not containing H₂SO₄ photolysis. Since this is obviously not the case, I wonder why they did not show all results including H₂SO₄ photolysis right away. Second, I am even more surprised about their result, namely that H₂SO₄ photolysis is completely negligible. Because this contradicts my statement of a "massive" effect, I would have expected them to discuss possible reasons for this contradiction. As my statement referred to background conditions (non-SAI), we reran our CCM (SOCOL) with SAI and found that the large impact reduces to <10% in sulfate concentration in the center of the aerosol layer. This confirms the figure shown by the authors. Now, that they have demonstrated that H₂SO₄ photolysis indeed plays a negligible role under SAI conditions, I do not understand why the authors continue to write "Note that the photolysis of H₂SO₄ gas ... is not included in the model." Unless readers refer to this review, they may ask themselves the same questions I did.

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I agree with publication in ACP, provided that the misleading statements regarding the aerosol formation are corrected and the consequences of neglecting H₂SO₄ photolysis are mentioned.