

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

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Review by Thomas Peter

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 $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ droplets proposed for SAI formed at the stratopause, even though temperatures at these altitudes are so high that the formation of $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ 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 H_2SO_4 , 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 $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ droplets in their model form at the stratopause was a misunderstanding. The “Results” section now states: “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.” 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 SO_2 to higher latitudes, from where the SO_2 (plus some already formed gaseous H_2SO_4) is transported further to lower altitudes, finally oxidized completely to H_2SO_4 and forming $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ droplets only below an altitude of 30–35 km (through bimolecular homogeneous nucleation or heterogeneous nucleation, e.g., on meteorite dust particles).

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 NO_x 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.

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.

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.