

The paper by Axebrink et al. highlights an important aspect for simulations of the sulfate aerosol evolution in the stratosphere after a volcanic eruption. They performed simulations with three different SO₂ emission profiles and show that the results depend strongly on the profile used.

Emission rate, altitude, profile and timing are important for a realistic simulation. The evolution of the sulfate aerosol formation is very non-linear and depends on SO₂ and background concentrations. The emission altitude determines the direction in which the volcanic cloud is transported in the model. Many of these aspects are nicely highlighted in the paper.

The paper is well written and reads well. I recommend publication after a few minor corrections.

Thank you for helping us to improve the manuscript. Your insights and comments on the microphysics clearly helped us to improve the interpretation of the simulations and their relation to the satellite observations.

Please see the answers to the reviewer's comments below.

General:

The article misses the aspect of microphysical processes and the consequent differences in particle size between the different simulations. Scattering of sulfate aerosols depends on the particle size (e.g. Laakso et al 2022). This could be another reason for the differences in the simulated forcing. My recommendation is to take a look at the particle size, e.g. calculate the effective radii, and include a short discussion.

We agree that the microphysical processes are of great importance for the climate impact of the stratospheric sulfate aerosol. We have added a figure of aerosol effective radii and one displaying the AOD divided by stratospheric sulfate mass to investigate the importance of the size distribution for the light reflection. In relation to this figure (new Figure 8), we have included a paragraph discussing the microphysics importance for aerosol light scattering and the differences between the simulations. See section 3.3.

The description of your experiments is a bit misleading. The table implies that your vertical grid of the S21s simulations has a vertical grid spacing of 200 m. This is not the case. Instead, you must interpolate the input data onto the model grid. Right? So it is not the resolution of the injection input data that is important. What is more important is the injection profile in the model. What does it look like and what does the model grid look like? Figure 6 shows quite similar profiles for June. So I assume that the height resolution of the input data disappears in the model. You can show and explain more details and show an initial phase profile.

We agree with the reviewer that Table 1 was misleading. We have changed one of the headings and the Table caption to clarify that the Table describes the input datasets. We have also added this sentence to the methods section: "When the SO₂ is emitted in the model it is interpolated to the model grid which is the same for all simulations."

When the datasets are read in by the model, they are interpolated to the model grid and it is true that the effects of the resolution of the datasets are reduced. Since the emissions using the different datasets are emitted over several days and at different days it is not possible to produce one single injection profile. Nevertheless, the vertical SO₂ profiles shown in Figure 3a are the total SO₂ in the model 5 days after the eruptions started, i.e. the first day that the SO₂ emissions in all simulations have been injected in the model and the first time at which the injection profiles from the simulations can be compared to each other. We have expanded the results section with regards to Figure 3a specifically focusing on the initial phase of the simulations.

A clear reason for the differences in the forcing calculations is the simulated altitude of the volcanic cloud (Fig 6). Surprising is the different behaviour between S21-1D and S21-3D. A better explanation should be added.

We have made more in-depth analysis of the differences between the simulations, including investigations of aerosol microphysics. Please the comments below.

Specific comments:

Line 38: What means significant here?

We have deleted that sentence to comply with remarks from reviewer #2.

Line 43: A residence time of several years is quite unrealistic.

We agree and deleted the word 'several'.

Line 49: No, the climate impact is estimated from the simulations. Satellites provide estimates on eruption rate and altitude.

We agree and changed the sentence to *"...Global modelers often use satellite-based observations of volcanic SO₂ as input when simulating the volcanic impact on the stratosphere and climate..."*

Line 62:climate impact. Do you have a reference?

Our statement is based on pure logic. Differences in wind directions at different altitudes and locations may produce large differences in the spread and stratospheric lifetime of the volcanic sulfur. We changed the sentence to: *"... Small errors in horizontal or vertical transport may cause errors in the evolution of the SO₂ distribution (Tilmes et al. 2023) and transport of the formed sulfate particles, and ultimately in the resulting climate impact..."*.

Line 63: Maybe, but due to the non-linear nature of the aerosol formation your simulations miss the early phase of the eruption. This may lead to different particle sizes and aerosol concentration.

This is indeed an uncertainty and complication of our approach. However, we find only small differences in particle size distributions among the three simulations in the first week after the eruptions (SO₂ injection to the model).

Line 72-73: Add a reference for the dataset.

We have added the reference to Mills et al. (2016).

2.2 Model description: Information on the vertical grid are important here. Table 1: Very misleading. Starting with 'Simulation name' and 'Vertical resolution' this could also be the vertical resolution of the model. What means single column? Should be in the model 0.95 x 1.25 as well.

Thank you for pointing this out. We agree that this was confusing. The table refers only to the input datasets and not the simulations. We have changed "Simulation name" to "Dataset name". We have also clarified this even more in the Table caption which now reads: "Properties for the three input SO₂ datasets."

Fig 1: b) shows the vertical integral, the burden, of SO₂?

Yes, this is the vertically integrated SO₂ in the S21-3D dataset. We have clarified this figure text also based on comments from Reviewer # 2. The figure caption now reads: "...*(a) Vertical SO₂ profiles for the three input datasets of each simulation. The vertical profile for M16 and S21-1D is the summed total injection for the eruption on the 15th and the 16th of June, whereas the vertical profile for S21-3D is the total injection on the 19th of June. (b) Vertically integrated total amount of SO₂ for the S21-3D dataset. The red triangle marks the location of the volcano Sarychev Peak. (c) latitudinally integrated total amount of SO₂ for the S21-3D input dataset. (d) longitudinally integrated total amount of SO₂ for the S21-3D input dataset...*"

Line 165: SO₂ does not disappear.

We agree with the reviewer. We have rephrased this sentence.

Line 184 and line 187-188: This argument is valid if your volcanic cloud stays at the same altitude (in meters). But from Fig 6 you can see a different behavior of the cloud. So the sulfur evolution cannot be deduced from Fig 1S alone. Microphysical processes are also important. You may have different particle size, different sedimentation, and different altitude, all of which affect the lifetime.

We agree that the sulfur evolution cannot be deduced from Fig S1 alone. The relatively small differences in particle sizes among the simulations cannot explain the differences in residence times. The S21-3D simulation results in the largest particles and shows the longest duration of aerosol perturbation in the stratosphere. Hence, sedimentation cannot explain the differences between the simulations shown in our Figures (e.g. Figure 6 and 7).

Line 189: The wording is difficult here. In principle, a model simulates one out of many possible weather/climate conditions. So, a model has its own reality. As you nudge the model, the results should be close to reality. On the other hand, a strongly nudged model cannot feedback and change the transport of the volcanic cloud caused by the heating inside the cloud due the absorption.

We agree with the reviewer. Our intention is point out the importance of the transport. We have modified the sentence to clarify this.

Figure 3: The difference between the solid and the dashed line is difficult to understand for the quick reader. My first impression was that the dashed line shows the total SO₂ (volcanic plus background) and the solid the volcanic SO₂ only.

The solid line shows the volcanic SO₂ in the stratosphere while the dashed line shows the total volcanic SO₂ (stratospheric and tropospheric). The background has been removed by subtracting the noVolc SO₂. We have changed the figure caption to make this clearer.

Figure 3: Do you show an average over a specific region or the max values or another mean?

These are globally summed values. We have added this information to the figure caption.

Line 194-195: I don't agree with this statement. As said before, in S21-3D the model misses the initial phase of the sulfur evolution and you get different results. A nudged model should simulate the transport well in case your timing and altitude of the injection is correct.

It is true that using the S21-3D dataset misses the sulfur evolution during the first 3-4 days. However, since the aerosol formation during those days occur under rather extreme conditions inside a concentrated volcanic cloud it is uncertain to what degree a global model can realistically simulate the aerosol formation during these days.

A nudged global model will simulate large scale features of air transport well but will still struggle to mimic mesoscale phenomena such as jet-streams due to the limited resolution of the model. This is of particular importance for volcanic eruptions that reach the midlatitude tropopause region. In the case of the Sarychev eruption, there was a rapid cross-Pacific transport of the volcanic SO₂ layers indicating that transport by jet-streams are important.

Line 197: Better first weeks.

We changed accordingly.

Figure 4: Units? Do you show Tg (SO₂) and Tg (SO₄)? The same unit should be used, which is Tg (S).

We have changed the figure according to the reviewer's suggestion. We have also updated the figure caption based on this.

Line 219: I don't understand the averaging of the 5th column. Do you show a global average? A zonal mean is an average over longitudes, so where is the difference to the other figures? Ahh, OK, the caption says NH average. Please, change the sentence and say NH average in the text as well.

Thank you for pointing this out. We have corrected this according to the reviewer's suggestion.

Figure 5: Do you show the vertical integral, the burden here? Why do you subtract SO₂ when you show SO₄?

This was a typo. It should say that we have subtracted the SO₄. This has been corrected.

Line 230: This differs between the months. In July M16 seems to be the best.

We agree with the reviewer on this and have modified the text to reflect this. The text now reads: *"Above these lowest altitudes, the model simulations have similar extinction coefficients as the CALIOP observations. During July, the M16 profiles bear most resemblance to the CALIOP profiles but after this month, the profiles from the S21 simulations have values more similar to the CALIOP observations."*

Line 239: OK, but the altitude is substantial. I would say in the S21 simulations, especially S21-3D, the aerosol is at a too high altitude.

The (small) difference between the S21 simulations and CALIOP could be the result of the aerosol being placed at a too high altitude (due to aerosol formation or transport) but another explanation is slightly too high extinction values in the S21 simulations. During August to October the vertical profiles of the aerosol extinction coefficient have similar slopes for the S21 simulations and CALIOP at 14- 20km altitude (Figure 6). The simulations show slightly higher aerosol load (aerosol extinction) than the observations.

The difference between the S21 simulations and the satellite observations may (to a large degree) be explained by uncertainties in the injected SO₂ mass. The S21 simulations overestimate the aerosol extinction coefficient values mostly in June-July. Although, the aerosol extinction coefficients differ between the S21 simulations and satellite observations, their difference is smaller than the typical uncertainty of volcanic SO₂ estimates. In the case of the Sarychev eruptions the SO₂ estimates ranges from 0.6 – 1.2 Tg. While the extinction coefficient values are non-linearly connected to the SO₂ mass, our new figure (new Fig. 8) shows only small differences in the AOD response to SO₂ (or rather AOD/SO₄) among the three simulations. Hence, large part of the difference between the S21 simulations and the satellite observations likely stem from uncertainties in the SO₂ estimates.

Line 249: Please, explain this with a few more words.

We have expanded this sentence.

Line 257: Agree, but your aerosol is not at the right altitude. Timing of the eruption is important as well.

It could be that our aerosol is at a too high altitude. But the difference between the simulations highlights the importance of the vertical placement of the SO₂ injections. We have changed the text to better reflect this and the sentence now reads: *“This large difference exemplifies the importance of the vertical placement of volcanic SO₂ injections in global climate models.”*

Figure 6: Please add the average area, e.g. zonal mean.

We have added that these are zonal means.

Fig 6e, are the colours correct? S21-3d show the smallest values and highest altitude.

Yes, the colors are correct. Since CALIOP and the simulations have different tropopauses and latitudinal extent the averaging in this figure is not done over all the gridboxes used in the other figures. Also, since the S21-3D data is injected days later than the other datasets, this profile is averaged with more days without Sarychev aerosol present.

Fig 7: global mean?

Yes, thanks for pointing this out. We have added this information to the figure caption.

Line 270: All three simulation reproduce the observations. M16 has a larger bias, an error of roughly 25%. This is a bit, but it could be much worse.

We agree that the M16 simulations could reproduce the aerosol extinction coefficients in parts of the stratosphere during some months and decided to delete this statement (the last part of the sentence).

Line 271 to 282: This last paragraph is right and important. However, your results are a bit more complex. I cannot agree that the S21 simulations are in general better than M16. S21-1D is better, but S21-3D simulates the volcanic cloud in a too high altitude. This impacts all results, especially lifetime and forcing.

It is true that the S21-1D and S21-3D simulations predict aerosol load at higher altitudes than the observed aerosol extinction coefficient values. On the other hand, M16 underestimates the aerosol extinction coefficients at all altitudes during Aug-Oct.

Line 275: The limits are more complex. You do not mention microphysical processes. They are extremely important and highly non-linear. The sulfur evolution depends on your emission profile, emission rate, and on the timing of the eruption. It depends also on the microphysical scheme (Tilmes et al, 2023; Laakso et al 2022). You have to broaden your discussion and you should also discuss your results more critically.

We agree that the sulfur evolution is limited by more factors than SO₂ vertical distributions alone. We have changed the sentence to clarify that the vertical SO₂ profile is one of the factors influencing

the sulfur evolution (which in turn impacts the climate). The sentence now reads: *“These findings highlight the need to produce high-vertical resolution datasets of volcanic SO₂ injections to the stratosphere and indicate that our present understanding of volcanic climate cooling is **in part** limited by the SO₂ profiles.”*

We have also added discussions regarding the microphysical processes to the manuscript. Please see our answers to comments above.

Specific comments:

Laakso, Anton, Ulrike Niemeier, Daniele Visioni, Simone Tilmes and Harry Kokkola, Dependency of the impacts of geoengineering on the stratospheric sulfur injection strategy part 1: Intercomparison of modal and sectional aerosol module, *Atmos. Chem. Phys.*, 22, 93–118, <https://doi.org/10.5194/acp-22-93-2022>, 2022

Tilmes, S., Mills, M. J., Zhu, Y., Bardeen, C. G., Vitt, F., Yu, P., Fillmore, D., Liu, X., Toon, B., and Deshler, T.: Description and performance of a sectional aerosol microphysical model in the Community Earth System Model (CESM2), *Geosci. Model Dev.*, 16, 6087–6125, <https://doi.org/10.5194/gmd-16-6087-2023>, 2023.