Quantifying the decay rate of volcanic sulfur dioxide in the stratosphere

Response to reviewers and editors in red. All line numbers refer to the revised manuscript.

Editor

Furthermore, please revise based on the following two comments:
line 41: "Once formed, stratospheric sulfate aerosols have a residence time of 1 to 2 years (Kremser et al., 2016)." First, this estimate of residence time is valid only for large tropical eruptions--many references will quote a different residence time for high latitude eruptions, which is relevant to your study since two of the eruptions you focus on are high latitude. Secondly, I cannot find this precise statement in the paper by Kremser et al. (2016), although it is common in other references.

Thank you for the comment. We have updated the language to be more precise and also added more appropriate references. Please see lines 43-46 in the revised manuscript:

> "Once formed, the residence time of stratospheric sulfate aerosols ranges from a few months to a couple years and depends on the latitude, injection height, and time of year of the eruption. High latitude eruptions with relatively low injection heights are associated with shorter residence times, whereas the aerosol cloud from tropical eruptions with high injection heights can persist for 1 to 2 years (Toohey et al., 2025; Myhre et al., 2013)."

line 224: Here you claim similarity between the decay timescale of SO2 and the timescale of the increase in sulfate aerosol mass, but the latter seems that it would be very sensitive to the period chosen to perform the fit over. How did you choose the particular period for the fit, and how sensitive is your conclusion to the choice of period?

Thank you for the comment. We have elaborated on this comment in Section 3. Please see the revised manuscript (lines 230-255).

Reviewer #1

l. 227 (of the file egusphere-2024-3525-manuscript-version3.pdf), 'high spectral resolution':

I'm convinced that this should read 'reduced spectral resolution' since, as it is correctly stated in line 125: 'the spectral resolution was 0.0625 cm−1' and this is the reduced spectral resolution of MIPAS which is valid for the years 2005-2012.

Thank you for bringing this up again, and you are most definitely correct. We apologize for the mistake! The wording has been fixed in the newest version of the manuscript.

Reviewer #2

Review of "Quantifying the decay rate of volcanic sulfur dioxide in the Stratosphere"

This is my second review of the paper. A number of my recommendations were ignored in this revision.

Thank you for taking the time to review the paper again. The authors would like to clarify that we addressed all of your initial comments during the first round of revisions. These are contained in egusphere-2024-3525-author_response-version2.pdf (found in the MS records), as well as in the interactive discussion section. Furthermore, based on the line numbers referenced in the reviewer's comments, it appears the reviewer read the initial submission of the paper as opposed to the revised one.

There are two main issues that need to be addressed.

(1) The authors fail to resolve or even suggest a resolution for the very different SO2 decay rates between total column SO2 (OMI) limb SO2 (MLS/MIPAS) (see Table 2). They need to come up with a plausible explanation for these large differences otherwise this paper is adding noise (not signal) to the issue of SO2 decay.

The authors believe this text in Section 4.3 addresses this concern (lines 435-440):

> "There are a couple of possible explanations for this. While the OMI data used here is designed to give an estimate of SO_2 mass in the stratosphere (Sect. 2.1.3) there is potential for tropospheric SO_2 to influence this measurement. Tropospheric SO_2 will generally get removed much quicker than that in the stratosphere, and could be

skewing the decay rates reported here. Additionally, there is a known bias in the OMI data due to the limited sensitivity of nadir instruments as the plume disperses (see Sect. 2.1). Both of these should be considered more carefully when analyzing OMI SO_2 following an eruption."

Additionally, we have added the following in the conclusion (lines 534-536):

> "This is a bias, perhaps due to interference from tropospheric SO_2 and nadir instruments' limited sensitivity to dispersed plumes. It should be considered when analyzing volcanic SO_2 with OMI and other nadir-sounding instruments."

(2) I also have a serious comment on the interpretation of the exponential decay of SO2 based on the recent paper by Toohey et al. (2025; https://doi.org/10.5194/acp-25- 3821-2025). SO2 can decrease due to two processes in the 10-22 km region: (1) conversion to sulfate aerosol and (2) transport out of the stratospheric domain. These processes have different time scales. The closer the eruption is to the troposphere boundary, the dynamical transport will accelerate the loss - assuming that SO2 lifetime in the troposphere is much shorter than in the stratosphere (about 2 weeks, Beirle et al., 2014). This dynamical effect will appear as a faster exponential decay which the authors attribute to stratospheric chemistry. To give an example, let us say that an eruption takes place close to the tropopause – if the gas were neutral in the stratosphere and the loss was only in the troposphere, the stratospheric lifetime might be ~2 months. If we fold in the SO2 actual chemical decay time in the stratosphere (say 1 month McKeen et al., 1984) then the observed lifetime would be ~20 days. In other words, the observed lifetime of SO2 is a mix of chemical and dynamical lifetimes whereas here it is interpreted as purely chemical.

Thank you for this comment. However, the authors argue that vertical transport will be a higher order effect on the decay rates calculated here and thus only have a minor impact. For instance, the lifetime of sulfate aerosols formed after volcanic eruptions is on the order of several months to a couple years (Toohey et al., 2025, Myhre et al., 2013), which is appreciably longer than the timescales for SO_2 oxidation by OH; these aerosols are removed not only by vertical advection, but also sedimentation (which results in a faster aerosol removal than could be done by advection alone). Thus, we believe (in agreement with previous literature) that the variation of decay timescales with height are primarily due to chemical processes.

To hopefully clarify some of the confusion around the role of vertical advection, we have added the following discussion to the paper (lines 371-376)

> "Vertical transport by the background circulation of the stratosphere is unlikely to have a significant impact on our results as it is quite slow---on the order of tenths of mm/s or hundreths of km/day---compared to the timescale of SO_2 decay (Butchart, 2014). Khaykin et al., (2022) did report an unusual radiative self-lofting of the Raikoke volcanic plume in 2019; the observed vertical ascent for this eruption was upwards of 2 mm/s

(0.17 km/day) and would be fast enough to impact our results. This phenomenon has not been noted for any of the volcanoes examined here, though it is a potential source of uncertainty and worth examining in future work."

Specific comments are below.

Ln 12 It would be useful to the reader to indicate the range of lifetimes rather than leave it as "difficult to attribute"

Thank you for the suggestion. We have changed the text to read:

> "While the typical decay timescale for SO2 is on the order of a few weeks to a month, we find that uncertainties across different altitudes and eruptions results in lifetimes that can vary by more than a factor of 2. This makes it difficult to attribute variations in decay timescale to specific SO2 removal processes for the events examined."

Ln 30 'once the plume reaches the stratosphere'

We have changed the wording as suggested.

Ln 41 There a lot more volcanic aerosols impacting climate references (see Robock, Stenchikov, etc.)
In the literature review, you should cite McKeen et al. (1984) here as well as in line 173

We added references to Robock (2000), Stenchikov et al., (2009) to the statement about aerosols impacting climate. Additionally, we added the McKeen et al., (1984) citation to the recommended location.

Ln 90 Is there a reason TROPOMI wasn't used? Its spatial resolution is higher than OMI?

TROPOMI data begins in 2018 and thus doesn't cover the eruptions that we focus on in this paper, whereas OMI does. Furthermore, while TROPOMI does have higher resolution, we choose to focus on OMI as there were several papers that use OMI to derive SO_2 decay rates that motivated this paper initially (see, for example, Zhu et al., (2020), Zhu et al., (2022), and Krotkov et al., (2008)).

Ln 107 Livesey is spelled wrong.

Thank you for the catch! The spelling has been corrected.

Ln 112 Actually, OMI does provide ozone profiles using the SBUV profiler methodology so what you say here is not quite true.

We have updated the text to specify that OMI does not provide high-resolution vertical resolution for SO_2.

Ln 115 The Dobson unit definition is wrong (cm^{-2})

Thank you for pointing this out. The unit has been corrected.

Ln 151 I asked this question in a previous review, why not divide that atmosphere from 10- 14 km, 14-18 km and 18-22 km. The lower bins include the troposphere in the tropics which is a quite different region than the stratosphere.

This is an important point, and one that we addressed in our response to your initial review of the manuscript. The line number referenced does not align with the location of this discussion in the revised manuscript, which perhaps suggest that our response was missed. Since we have already addressed this point, I have copied the previous response below:

Thank you for the comment. The main rational behind using the three layers is that it allows for comparison to previous work by Höpfner et al., (2015). We agree, however, that whether or not these layers fall in the stratosphere is going to be highly latitude dependent. As such, we have included a more detailed discussion of where these layers fall as a function of time-of-year and latitude in the beginning of Section 4.3 (lines 354 to 359 in the revised manuscript):

> "In particular, the tropopause in the tropics during the local summer is around 16 km, whereas that for the high northern hemisphere latitudes is closer to 11km (Hoffmann and Spang, 2022). As such, the majority of the three layers considered in this analysis are likely to be in the stratosphere for the Kasatochi and Sarychev eruptions. After the Nabro eruption, likely only the 18 to 22 km layer was initially fully in the stratosphere; however, the plume was quickly advected to higher latitudes—where the tropopause is lower—by the Asian Monsoon anticyclone in just a few days (Clarisse et al., 2014)."

We think that in order to facilitate comparison with past work, keeping the analysis focused on these three layers is the best choice. However, we have added a comment on the validity of this choice in light of the concerns you raise. See the updated text in Section 2.3 (lines 193 to 196) in the revised manuscript):

> "Given the variation in tropopause height with latitude, the 10 to 14 km and 14 to 18 km layers won't necessarily be entirely in the stratosphere in low latitudes (Hoffmann and

Spang, 2022). However, we use the vertical divisions here for consistency with past work, and future work could consider a division based on tropopause height."

Ln 198 Aura is not in a decaying orbit. To preserve fuel, Aura is not following Aqua and so the crossing time has drifted. With current fuel reserves, Aura (and MLS) will last until 2028 after which power levels are too low to run the instrument.

We have refined the language to read as:

"The Aura satellite, which carries the MLS instrument, is expected to last until 2028, while MIPAS operated from 2002 through 2012."

Ln 218 Do the V2 MLS SO2 retrieval problems persist in V5? See more about this below.

Yes, based on our Figure 3, for example, it appears similar problems with regards to the seasonal cycle persist in V5. See the following comment for more details and modifications that we have made to the text.

Ln 236, 243 The MLS data quality document clearly discusses the SO2 changes from early versions. There is no mystery here. The MLS user guide can be downloaded from the JPL website. Negative mixing ratios are an artifact of the retrieval. They are not bad data, and indicate that averaging over a larger volume is required.

We have downloaded and read through the MLS data quality document thoroughly. The changes stated in the data quality document for V5 mention changes in the channels used for O_3 and CO lines, which "will have secondary impacts on SO_2." Additionally, the documentation highlights that all versions of the MLS dataset are biased high "due to systematic errors in the MLS measurement system." There is no mention of the seasonal cycle or the change in magnitude between V2 and V5 that we report here in lines 308-310.

Furthermore, while the documentation does indeed mention that negative mixing ratios are a by-product of the retrieval, our results here show that these negative biases persist even when the data is aggregated over a large area (40°N-90°N, for example).

We have modified the text in lines 277 to 280 to make the reference to the documentation more explicit:

"Negative mixing ratios are unphysical and an artifact of the MLS retrieval algorithm; nonetheless, the MLS documentation recommends including them in subsequent calculations (Livesey et al., 2022). However, aggregating these negative mixing ratios over a large geographic area (e.g., 40N to 90N) and converting to mass results in large negative values that complicate the interpretation of the data, but are nonetheless included here."

We have also added some additional context to the MLS dataset in section 2.1.2 (lines 143 to 153):

> "We use Level 2 Version 5 (V5) daily swath SO_2 mixing ratio data, accessed at https://disc.gsfc.nasa.gov/datasets/ML2SO2005 (Read and Livesey, 2021). The V5 data features minor changes from previous versions, including improved cloud detection, changes in the calculation of O_3 and carbon monoxide (stated to have secondary impacts on SO_2), and updates to the handling of background radiance signals (Livesey et al., 2022). This data is obtained via the 240 GHz radiometer on the MLS instrument (Pumphrey et al., 2015). In addition to the SO_2 mixing ratio, this dataset reports the temperature at each pressure level, and we use this in our calculation of SO_2 mass and altitude above sea level.
>
> The MLS documentation highlights that the retrieval algorithm can generate negative mixing ratios, and the correct way to deal with these is to average over a sufficiently large horizontal area (Livesey et al., 2022). We apply all of the suggested masking for the data given in Livesey et al., (2022), and we average our data over 10° latitude bands. Even after masking and averaging, negative mixing ratios are prominent in the MLS data, particularly lower in the atmosphere."

Ln 250 I don't understand why you think that the seasonal cycle is unrealistic. Transport from the troposphere to the stratosphere has a seasonal cycle so I expect that upward transport of OCS and total SO2 would have a seasonal cycle.

It is not so much that the seasonal cycle is unrealistic, but the amplitude of the of the seasonal cycle seen here (~100 $GgSO_2$), particularly in the lower height bins, is too large to be explained by OCS transport or any other reasonable source of SO_2 in the stratosphere. Data from Höpfner et al., (2013) shows background SO_2 in the lower stratosphere on the order of only a few ppt (their figure 7), whereas the seasonal cycle of SO_2 from MLS has an amplitude on the order of ppb (see also Pumphrey et al., (2015)), which further suggests the seasonal cycle is unrealistic.

We have added some clarifying language about the MLS seasonal cycle in lines 284 to 289:

> "Additionally, the MLS mass in the 10 to 14 km and 14 to 18 km bins feature a seasonal cycle with an amplitude much larger than what is expected for background stratospheric SO_2 (Pumphrey et al., 2015; Höpfner et al., 2013). The background SO_2 values in this region of the stratosphere are on the order of a few tens to a hundred ppt (see Fig. 7 in Höpfner et al., 2013), whereas the seasonal cycle shown here and in Pumphrey et al. (2015) have an amplitude on the order of ppb. Furthermore, the amplitude of the MLS seasonal cycle is too large to be explained by other potential sources of stratospheric sulfur such as the annual flux of OCS into the stratosphere (Karu et al., 2023)."

Table 1. It would be helpful if you added latitude, longitude and exact eruption date next to the volcano name. If you ignore the 10-14 km range, where the decay rate may be accelerated due to dynamics, the numbers are in reasonable agreement. Also, the uncertainty of the MLS measurements is much higher between 8 and 12 km. The high uncertainty value for NABRO MLS SO2 seems a little weird to me. The 14-18 range is in the upper troposphere so I am not sure you aren't getting good data.

Thank you for the suggestion. We have added dates and geographic coordinates to both Tables 1 and 2.

As you highlight below, the MLS Nabro data is quite noisy, which leads to the large range for the 14-18 km decay rate reported in the table.

Fig. A2 What is producing the spikes about day 50 and 60? Those spikes are absent from MIPAS data.

It is not clear to the authors what these spikes are. Given that the explosive activity from the 2011 Nabro eruption had ceased by this time (see the eruption timeline from the Smithsonian Global Volcanism Program), we simply treat these spikes as noise in the signal.

There were a few other notable eruptions in 2011 after Nabro. Soputan erupted in Indonesia in early July of that year. Etna (Italy) and Hudson (Chile) erupted in October 2011. However, the timing of these eruptions is such that they cannot explain the spikes seen in the figure.

The increase in e-folding times with height found you found and also in Höpfner et al. is, I believe due to dynamics – transport across the tropopause is weaker at higher altitudes. This issued is mentioned in the beginning of the review.

Please see our response to comment (2) above.

Ln 320 You should fix line 112 to be consistent with this statement.

We have changed the wording to read (lines 422-423):

> "OMI reports the vertical column density of SO_2 and lacks the explicit vertical resolution for SO_2 provided by MLS and MIPAS."

Ln 330 Another explanation for the differences between OMI and limb sounders is that OMI is losing mass due plume dispersal. As the plume spreads out, the pixels with smaller amounts of SO2 will no longer register and thus the plume would "appear" lose SO2 when it is (in fact) not – the plume edges have fallen below the detection limit. The limb sounders also face this problem if they fail to acquire a plume on successive orbits, but since a significant amount of the OMI plume is in the

upper troposphere it is likely worse.
We have added the following text in lines 435 to 440 in the revised manuscript:

> "There are a couple of possible explanations for this. While the OMI data used here is designed to give an estimate of SO_2 mass in the stratosphere (Sect. 2.1.3) there is potential for tropospheric SO_2 to influence this measurement. Tropospheric SO_2 will generally get removed much quicker than that in the stratosphere, and could be skewing the decay rates reported here. Additionally, there is a known bias in the OMI data due to the limited sensitivity of nadir instruments as the plume disperses (see Sect. 2.1). Both of these should be considered more carefully when analyzing OMI SO_2 following an eruption."

Table 2 The large SO2 differences between OMI and the other instruments for Kasatochi is not explained and needs to be.

The authors also find this odd, and we are not sure why there is such a large difference for Kasatochi when compared to the other eruptions analyzed. We did modify the language in lines 490 to 492 to address this:

> "We also note that the MIPAS and MLS SO_2 masses in some eruptions are significantly lower compared to the Carn (2024) values. Whether this reflects fractional stratospheric inputs or biases due to limitations of sampling by limb-sounding instruments would be a subject for future research."

Ln 384 The version of MLS you are using, I believe, corrects for HNO3 and O3 interference. You need to reference and discuss the MLS V5 documentation of the SO2 retrieval.

We added some relevant comments and references to the MLS documentation to Section 2.1.2 (see lines 143 to 153 in the revised manuscript):

Ln 398 We are left hanging on the OMI vs MLS differences in decay rate.

Please see the reply to comment (1) above.

Ln 418 Please re-label HTHH as Hunga consistent with the community recommendation.

Based on the previous review, we have already changed this wording as suggested.