Quantifying the decay rate of volcanic sulfur dioxide in the stratosphere

Response to reviewers and editor in red

Note: An additional figure and section were added, and figure and section numbers in our responses refer to those in the updated manuscript.

Reviewer #1

l.22: '…is produced naturally in seawater':

This is only one of the several sources of OCS. It is even not clear if the major source is direct emission from sea-water, or if OCS is mainly produced by oxidation of CS2 and DMS (which originate in sea-water) (e.g. Kremser et al., 2016). Please be either more specific (or more general) here.

We have updated the language to be more specific as follows (see lines 22-25 in the revised manuscript):

> "An important source of stratospheric SO_2 in non-volcanic conditions is the photolysis of carbonyl sulfide (COS), which is the most abundant sulfur-containing gas in the atmosphere (Kremser et al., 2016). Important sources of COS include its direct flux from the ocean, oxidation of marine-originating dimethyl sulfide and carbon disulfide, and direct and indirect anthropogenic emissions, among others (Kremser et al., 2016)."

l.81: '…they potentially provide greater sensitivity to volcanic SO2'; l.86: 'the greater sensitivity of limb sounders may be advantageous'; l.332: 'This is likely to be a bias in the OMI data, perhaps due to the limited sensitivity of nadir instruments as the plume disperses…'; l.399: 'This may be a bias…'

I acknowledge the scientific caution when attributing biases in stratospheric SO_2 decay timescales to nadir-viewing instruments. However, the evidence presented in this study, as well as in previous works, strongly supports the existence of such a bias. This bias arises from the limited sensitivity of nadir-viewing instruments to diluted stratospheric SO_2 amounts compared to limb-viewing instruments, which benefit from significantly longer optical path lengths (several hundred times greater) through the dispersed plume. Given the robustness of this evidence, I recommend adopting clearer

language to describe this phenomenon. Such clarity is important to avoid potential misinterpretation of nadir-viewing data, particularly in the future, when a substantial volume of nadir-derived data remains available while limb-measurements might no longer be conducted.

This is a great point, and we have adjusted the language throughout the manuscript for increased clarity. For example, the text:

"This is likely to be a bias in the OMI data, perhaps due to the limited sensitivity of nadir instruments as the plume disperses"

has been changed to:

> "Additionally, there is a known bias in the OMI data due to the limited sensitivity of nadir instruments as the plume disperses" (see lines 401-402 in the revised manuscript).

l.98: 'high spectral resolution'

Should this not read 'high spatial resolution'?

When downloading the data from https://imk-asf-mipas.imk.kit.edu/mipas/, we had to select either "V5 high spectral resolution" or "V5 low spectral resolution". We used the V5 high spectral resolution data.

l.106: 'are reported on pressure levels with an approximate spacing of…'

The reported retrieval-grid of remote sensing data is generally not equal to its spatial resolution. Therefore, I would suggest to add the information on resolution from here: https://mls.jpl.nasa.gov/data/v5-0_data_quality_document.pdf, p. 157

Thank you for the suggestion. We updated the resolution numbers and added a citation for the data quality document. See lines 134-136 in the revised manuscript.

l.146: 'to the pressure coordinate of the MIPAS data'

Shouldn't this read 'to the altitude coordinate…'?

Yes it should! Thank you for the catch. We changed the text as suggested.

l.236: 'The reason for this remains unclear, and no explanation or documentation for this difference was found in the literature.'

You might try to contact the MLS-team for a possible explanation(?)

We have had personal correspondence with Hugh Pumphrey, one of the leading scientists working with MLS. He was also perplexed by the difference, and we did not get a resolution talking with him.

Chapter 3.2: 'Background seasonal cycle in the MLS data'

Is there any possibility to infer the disturbing seasonal cycle from comparing different years with less volcanic influence?

Thank you for the question and suggestion. At each day of the year, there is a spread of around 100-200 Gg in the SO2 seasonal cycle during non-volcanic times. Supposing we take the mean of the background years as the reference seasonal cycle, the actual disturbing seasonal cycle could be $\pm$50-100 Gg offset from the reference. This would impart a large bias on the calculated decay rate, thus limiting our ability to constrain the uncertainty.

The interfering background seasonal cycle is interesting in its own right, and to our knowledge under-explored in the literature. Given that it is likely due to O_3 and HNO_3, perhaps one might be able to calibrate the seasonal cycle based on observed values of these trace gases. However, we feel this would be beyond the scope of the current study. Nonetheless, we have added further discussion on this point (lines 305-312) in Section 4.2 as well as a supplementary figure (S2).

> "The approach outlined above samples possible seasonal cycles using the observed time series in a given year of an eruption. One could also potentially infer the interfering seasonal cycle from years without large volcanic eruptions; however, this presents its own challenges. There is a 100 to 200 Gg spread in SO_2 mass on any given day of the year in non-volcanic conditions (Fig. S2). Using the mean across years for the background seasonal cycle could result in an offset of $\pm$50 to 100 Gg from actual disturbing seasonal cycle. This difference is large enough to impart a significant bias in the estimated decay rate, thus limiting our ability to constrain the uncertainty. The seasonal cycle and inter-seasonal variability in MLS SO_2 is interesting in its own right: it warrants further investigation and could potentially be calibrated based on the observed amounts of O_3 and HNO_3 but this is beyond the scope of the current paper."

l.290, Table 1:

I strongly recommend consolidating all the information from Table 1 and Table B1 into a single comprehensive table. Additionally, I suggest including the uncertainties reported in Höpfner et al. (2015) and the results from the WACCM simulations, including those for the 18–22 km layer if available. This enhancement would greatly improve the readability of the discussion in Chapter 3.3, allowing readers to follow the analysis more easily without needing to consult multiple tables.

Thank you for the suggestion. We agree this improves readability, and Table 1 and the surrounding discussion in section 4.3 has been updated accordingly

l.290: 'Their values show a clear increase of e-folding time with height, which is not as apparent in our results.'

However, the decay timescales provided here also show increasing values (with one exception of MLS in case of Kasatochi). Further, I suggest also to try to provide best-estimates from your analysis of the decay-timescales at 18-22 km for MIPAS in case of Kasatochi and Sarychev – when looking at Fig. 2 and Fig. A1, there seems to be a clear signal.

We adjusted the text following Table 1 to read:

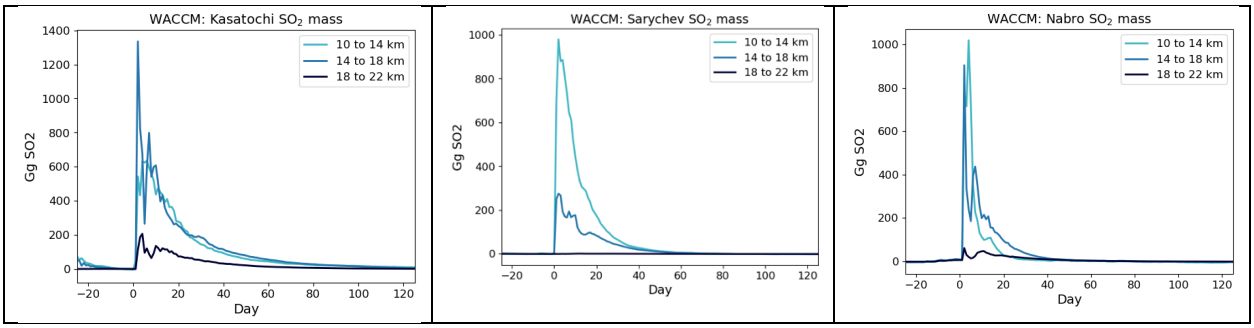
> "Their values show a clear increase of decay rate with height, which is generally also seen in our results (with the exception of some of the values for Kasatochi)" (lines 349 to 351 in the revised manuscript).

We also added MIPAS estimates for Kasatochi and Sarychev in the 18-22 km height bin in Fig. 3, Fig. 5. Fig. A1, and Table 1.

l.295-314: comparison to WACCM

The discussion may give the impression that the uncertainties in the decay timescales derived from the measurements are too large to allow for meaningful comparisons with the model. However, upon examining Figures 5, C1, and C2, it appears that for the Sarychev and Nabro eruptions, the model significantly underestimates the decay timescales compared to the limb-sounding datasets, whereas this discrepancy is less pronounced for the Kasatochi eruption. Could you provide possible explanations for this observed difference?

One possible explanation for the observed difference could be the fact that the model puts most of the SO_2 from the Kasatochi eruption into the 14-18 km height bin, whereas most of the SO_2 for the Sarychev eruption is in the 10-14 km height bin. The model also puts a substantial amount of SO_2 in the 10-14 km height bin for the Nabro eruption. As OH decreases with height in the model, SO_2 is going to get oxidized and removed more slowly the higher up it is. This could account for the slower decay timescales (and thus closer to MIPAS) reported for Kasatochi (as compared to Sarychev and Nabro) when looking at the larger vertical column. We added a figure (S2) to the supplement showing this and a brief discussion in lines 405-409 in the revised manuscript.



l.355: 'our main focus here is on the decay times of the stratospheric inputs of the indicated eruptions and not the total stratospheric mass.'

The entire chapter 5 of the manuscript (as well as section 2.5 'Calculation of total stratospheric SO2 burden') is dedicated to the 'Estimating the stratospheric SO2 burden'. Therefore, I don't understand this statement. I would suggest to delete this sentence and extend chapter 5 a bit by extending Table 2 to include the estimations of stratospheric SO2 mass by the extrapolation methods used by Pumphrey et al. (2015) and Höpfner et al. (2015). I would also suggest to add the results (M(t0)) from the fits

performed in the present work. It should be made clear that the method described here in section 2.5 is not adequate to calculate the total stratospheric SO2 burden in case of MIPAS and, to a less extend, also for MLS.

We have adjusted the sentence you highlighted to read (lines 465 to 466 in the revised manuscript):

> "The main focus of the paper is on the decay times of the stratospheric inputs from the indicated eruptions, and Table 2 explores the implications of such information for estimating the total stratospheric mass burden."

Your comment about presenting the inadequacy of method presented in Section 2.5 is well taken, and we have adjusted the text in Section 6 to emphasize this more clearly. For example, see lines 428-430 in the revised manuscript. Note we have removed Section 2.5 and included its content in Section 6.

> "As discussed in more detail below, this method proves inadequate for accurate mass burden estimations. This is particularly true for MIPAS, as the spectral bands measured by MIPAS saturate at high SO_2 concentrations. The inclusion of it in this paper provides a contrast compared to previously published total burden estimates derived from SO_2 decay timescale estimates…"

Finally, we have reworked Section 6 and updated Table 2 to include our own exponential fit-derived estimates of the total SO_2 burden. Please see the revised document for the changes.

l.400: '…and should be considered when analyzing volcanic SO2 with OMI'

I would suggest to add here: 'and other nadir-sounding instruments'.

Thank you for the suggestion; this text was added.

l.413-417: eruptions with ash

The eruption of Puyehue in June 2011 was also rich in ash. Have you tried to inspect that one for any effects on SO2 lifetime? (e.g. Griessbach et al., 2016, doi:10.5194/amt-9-4399-2016)

Thank you for the comment, as it is a good point. We considered other eruptions during the time period of overlap between MLS and MIPAS. However, we focus here on three specific eruptions in this paper because they were large enough to allow for a calculation of the e-folding time of SO_2. Puyehue, as well as other notable eruptions during this time period such as Grimsvotn had too weak and noisy of a signal, particularly in the MLS data, to calculate the decay rate. As we are interested in comparing MLS and MIPAS, we leave these smaller eruptions out of the analysis.

l.424: 'Our work suggests that the current SO2 data reported by available observational products are subject to significant uncertainty when examining the stratospheric lifetime of volcanic SO2 and suggests that more precise data is needed if chemical mechanisms and SO2 mass loading following an eruption are to be elucidated using observed decay times.'

On one hand, I support this statement, particularly considering the imminent loss of limb-sounding capabilities for stratospheric SO_2 observations, which will create a significant gap in our ability to monitor the stratosphere. On the other hand, I find the statement somewhat overly general. As noted in the manuscript, each observational technique has specific advantages and limitations in quantifying stratospheric SO_2. Therefore, to effectively evaluate and refine models, it may be more appropriate to tailor comparisons to align with the strengths of each dataset. For example, model results could be compared directly with nadir and MLS data closer to the eruption time, while comparisons with IR limb-sounding datasets might be more suitable for periods several weeks after the eruption.

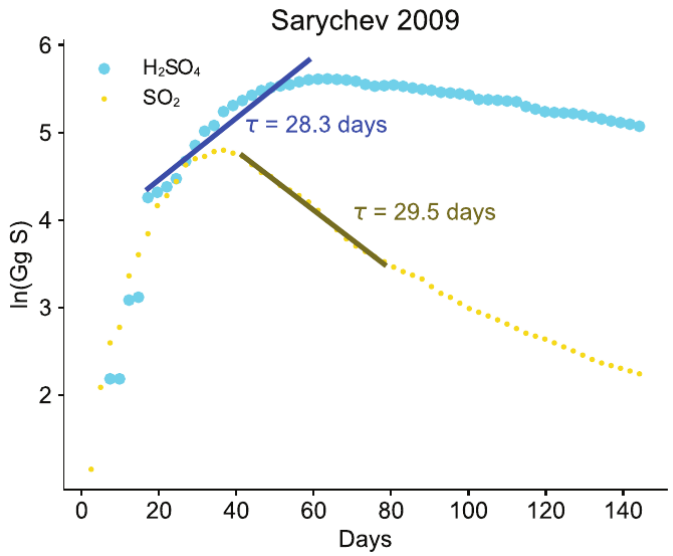
Thank you for the suggestion. We have modified the last paragraph (lines 523-527 in the revised manuscript) to read as follows:

> "Our work suggests that the current SO_2 data reported by available observational products are subject to significant uncertainty when examining the stratospheric decay of volcanic SO_2. The varying strengths and shortcomings of the different observational products should be accounted for when using them to determine chemical mechanisms and SO_2 mass loading. Furthermore, the forthcoming loss of MLS (the only limb-sounding SO_2 instrument in operation) will leave a significant gap in our ability to monitor the stratosphere."

Reviewer #2

I was hoping to see a 'back of the envelope' check against the total aerosol eruption mass burden – the end product of SO2 oxidation. This would be a useful addition. (See ***Schulte et al., 2023,https://doi.org/10.5194/AMT-680 16-3531-2023 on computing the total mass).*** There are a number of stratospheric aerosol sources you can use – but probably GLOSSAC is the best. This sort of 'stupidity check' would confirm that the SO2 estimates agree with aerosol production - which is why we care about this.

Thank you for the comment and the suggestion. We have added short section (Sect. 3 in the revised manuscript) and an additional figure (Fig. 2 in the revised manuscript). Discussing this point and showing the similarities between the two timescales. The figure is copied below:



In regards to your suggestion about GLOSSAC, looking through the various GLOSSAC data products available, we only found data products available on a monthly resolution (https://asdc.larc.nasa.gov/project/GloSSAC). As we are interested in processes happening on the order of weeks, monthly resolution data won't have the necessary level of detail. However, using sulfate aerosol burden estimates from MIPAS (see Günther et al., 2018; https://doi.org/10.5194/acp-18-1217-2018), we did a rough estimation for the timescale of sulfate aerosol formation for the 2009 Sarychev eruption. This is using MIPAS data from 10.5 to 22.5 km. The ~28 day time scale of sulfate aerosol formation aligns well with our estimate of a SO_2 decay time scale for the eruption (25-30 days).

An additional point we'd like to make here is that detecting perturbations to the stratospheric aerosol layer is not trivial due to the constantly varying background (e.g., Solomon et al., 2011; DOI: 10.1126/science.1206027). This makes defining an appropriate baseline for calculating timescales associated with a perturbation challenging (and is a similar issue to that seen with MLS in our analysis.)

The MLS algorithm, as I understand it, generates negative mixing ratios for data on the edge of observability. The correct way to deal with these are to average the data over larger regions including both positive and negative mixing ratios. I was looking for a discussion of this and mention of MLS validation also found in Livesey et al. (2022) (found at https://mls.jpl.nasa.gov/eos-aura-mls/documentation.php). Discussion of how to use the data including quality flag screening that is appropriate for SO2 is also found there and should be mentioned in the data description (lines 65-75). An equivalent MIPAS discussion is needed.

We added the following text to expand on this detail of the MLS dataset (lines 139 to 143 in the revised manuscript):

> "The MLS documentation highlights that the retreival algorithm can generate negative mixing ratios, and the correct way to deal with these is to average over a sufficiently large horizontal area (Livesay et al., 2022). We apply all of the suggested masking for the data given in (Livesay 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."

We also added details for how we did the data masking based on the suggested guidelines for MIPAS (lines 128 to 129 in the revised manuscript):

> "As suggested within the MIPAS data files, we select valid data by only using points where visibility == 1 and akm_diagonal > 0.03."

The division of the SO2 into three separate regions (10-14), (14-18), (18-22) made me uncomfortable. At high latitudes in winter these regions are all in the stratosphere – in the summer the 10-14km may include the troposphere. In the tropics (as with Nabro and many other eruptions), only 18-22km is entirely in the stratosphere. This distinction can play an important part since the water vapor content and OH concentration (eq. R1)of these layers can be quite different – upper troposphere vs lower stratosphere – and thus will affect the decay rate. Since MLS and MIPAS also make water vapor measurements, the water vapor content can be added to the

analysis. It seems to me that the the authors should have used two layers - below the tropopause and above the tropopause - rather than what was done here. It is easy to get tropopause height information from reanalysis data sets (GFS, MERRA2, ERA5).

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 318 to 323 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 184 to 187 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."

The authors neglect the transport between the layers. Exchange between layers needs to be discussed as possibly influencing the decay rate.

We had considered the potential role of transport in influencing our decay rate but came to the conclusion that vertical transport is too slow to significantly affect the decay rate. The processes of interest here are on the order of weeks, and at most we are using a window of 25 days in our calculations. In comparison, typical time scales associated with vertical transport in the stratosphere are on the order of tenths of a milimeter per second or hundreths of a kilometer per day (e.g., Butchart (2014); 10.1002/2013RG000448).

There is evidence in self-lofting of volcanic plumes, similar to that seen in wildfire plumes (e.g., Khaykin et al., 2022; https://doi.org/10.1038/s41598-022-27021-0). However, this has not been noted for the volcanoes analyzed here. Nonetheless, it is a potential source of uncertainty.

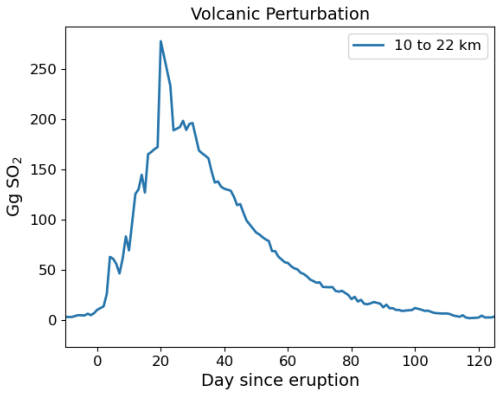
Both of these points have been elaborated on in the second paragraph of Section 4.3; see lines 334 to 340 in the revised manuscript.

> "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 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."

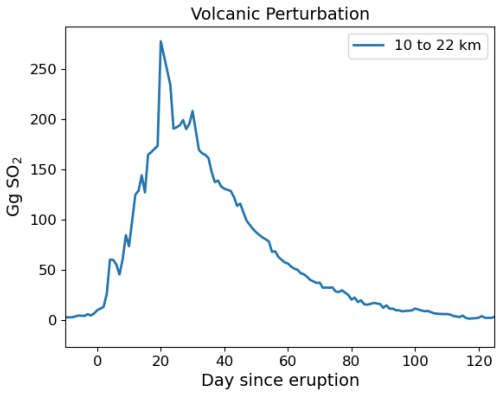
The authors are using a zonal mean SO2 on a 10° latitude grid (ln 153). It seems like they could also construct a tighter latitude grid (say 5°) and a longitude grid and select high SO2 regions which might reduce the uncertainty (e.g. Fig. 2). I would like to see how this affects their decay rate and agreement between the two satellite instruments.

In general, we find that using smaller latitude bands has almost no impact on the shape of the perturbation and the decay. We have included an example from the 2009 Sarychev eruption below. On the left is the time series of the MIPAS SO_2 perturbation calculated using 10° latitude bands, and on the right is that using 5° latitude bands. The two curves are nearly identical.

Left: 10° bands



Right: 5° bands



I don't think adding OMI SO2 helps the paper at all. In fact, it just adds noise, not signal. This is because the OMI total column includes massive amounts of tropospheric SO2 (for most eruptions) which – as the authors note – probably explains the significant differences in total SO2 mass and decay rate. If you add OMI you might as well take a look at SO2 measurements from AIRS (mentioned line 71) as well for completeness. Anyway, I suggest you just drop this section – it really adds nothing.

Thank for the comment. We are in favor of keeping the OMI section in the paper as we think that the comparison between MLS and MIPAS (both limb-sounding instruments) and OMI (a nadir-sounding instrument) helps identify some of the important discrepancies that arise between the two, particularly in light of some studies that have used OMI-based measurements as evidence for different SO_2 oxidation pathways (e.g., Zhu et al., (2020)). As you mention, there are other nadir-sounders we could have used, and indeed, a comparison between these and the limb-sounders analyzed here could be a useful thing to do in the future. For now, we think such a step is beyond the scope of the current paper. We adjusted language in lines 380 to 384 in the revised manuscript to better emphasize this:

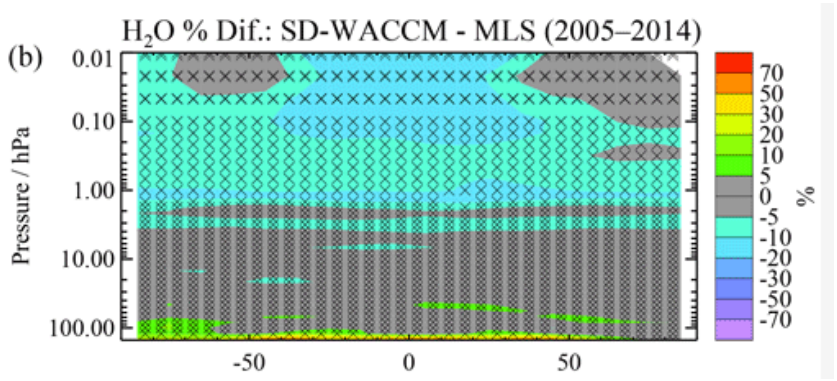
> "We compare the results from MLS, MIPAS, and WACCM with SO_2 retrievals from the Ozone Monitoring Instrument (OMI). OMI is a popular choice in recent work examining the decay of SO_2 following eruptions (e.g., Carn et al., 2022; Zhu et al., 2020; Krotkov et al., 2010), and we include an analysis of it here for a comparison of how limb-sounders and nadir-sounders capture the removal of volcanic SO_2. Note that OMI is just one of several nadir-sounding instruments that measure SO_2, and a detailed comparison with other instruments (such as AIRS) is left to future work."

Furthermore, one of the issues with OMI is it likely does contain tropospheric influence. This, combined with its known detection limit issues, are likely to contribute to a faster-than-realistic decay of SO_2, which is what we try to show by including it in this paper. We have updated the language in Section 5 to highlight this point (lines 399 to 403 in the revised manuscript):

> "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 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."

The WACCM simulation is interesting but barely discussed (Fig. 5). Take a look at the water vapor in WACCM. Was it the same as MLS observations? This might explain the accelerated decrease.

This is an interesting suggestion, as water vapor differences could very well be the culprit here. However, previous work comparing MLS water vapor to WACCM shows that there is generally very good agreement between the two in the region of interest for this paper. For example, see Figure 3 from Froidevaux et al., (2019); (https://doi.org/10.5194/acp-19-4783-2019):



We added a brief discussion about this in the second-to-last paragraph of section 4.3 (lines 362-365 in the revised manuscript):

> "There are a variety of potential causes for the discrepancy between the model and observations. One could be differences in water vapor and between the model and observations. However, comparisons between WACCM and MLS water vaper generally show strong agreement, and it is not clear that this should be the main culprit (Froidevaux et al., 2019)."

Minor comments:

Add layer labels to Fig. 2

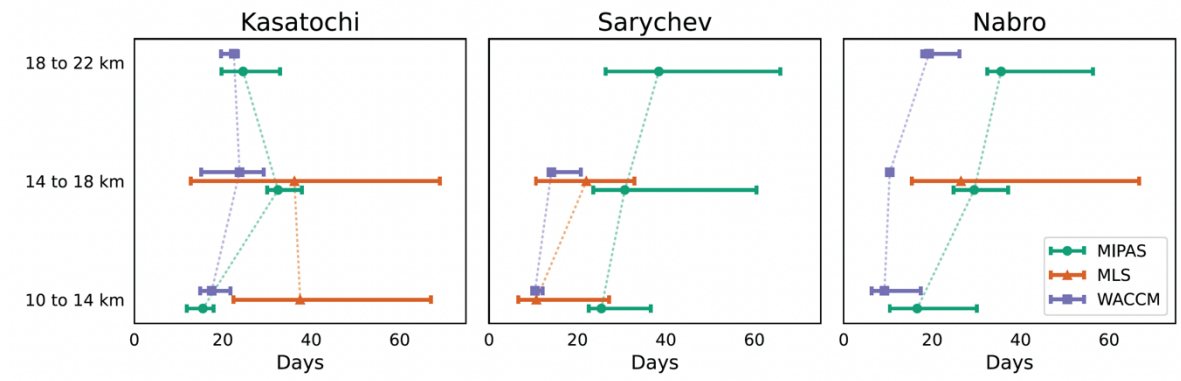
We think the legend sufficiently conveys which lines correspond to the different layers, and adding labels directly on the plot would add unnecessary clutter to the figure.

Line 287 'less uncertain' - how about 'better'

Thank you for the suggestion. We updated the text.

Fig 4. Why not connect the dots vertically. The figure – as is, is a little hard to read.

Our updated figure 5 is shown here and has been added to the paper.



Line 418 "Honga-Tonga" - the APARC group recommends using 'Honga' not HTHH or HT or other acronyms. The Honga eruption is a good example where hydrolysis probably played a critical role in accelerating the decay of SO2 and conversion to aerosols as noted. This is why I recommend the authors also take a look at H2O in other regions.

We changed the wording as suggested.

Editor #2

L50: This introduction of a result of this study is misplaced in the introduction. Please move to the results section.

We have moved this result to a new section (Sect 3, lines 220-226) in the results part of the paper. Additionally, we moved the relevant figure from the supplement to the main paper.

L62: "Do not tell the whole story" implies there is something new we will learn from MIPAS, but the sentence goes on to give a result which is consistent with the column measurements discussed in the prior sentence. Some different language would make this easier to understand.

Thank you for the suggestion. We have changed the sentence to:

> "However, total column measurements obscure vertical variations in SO_2 oxidation rates within the plume of a particular eruption: Hopfner et al., (2015) use vertically resolved observations from the Michelson Interferometer for Passive Atmospheric..." (lines 58-60)

- L84: “concentration” is a physical quantity that is not used much in this study, I wonder if mass would be a better example to use here?

Thank you for the comment. We have changed the word to “mass” in Section 1.1.

- L86f: I am fairly certain there is one too many derivatives here, and a missing negative sign. What you plot later and take the slope of is just ln(W(t)).

Thank you for the catch! We removed the extra derivative.

- L227, 229: Colloquially, we often talk about the impact of volcanos, etc., but formally it’s really the “eruption” that is important here, not the volcano.

We adjusted the wording as requested.

- L251: “elevation” should be “altitude”
We have adjusted the wording as suggested here and throughout the manuscript.

- One of the main conclusions is that uncertainty in the decay timescale “limits the ability to accurately determine the initial mass loading following an eruption when fitting applying an exponential decay to the SO2 data”. But you do not calculate the resulting uncertainty in the SO2 estimate. This is a clear hole in the argument that could be easily filled, since for every value of decay timescale, there will be a corresponding value of the injection amount. Instead, you provide a different type of injection estimate, based on the positive increases in the SO2 timeseries. But I find this estimate unconvincing, since it is clear that due to instrument and/or sampling issues, the timeseries are biased low in the first days when the timeseries apparently increases, but it is likely that SO2 is being removed through that period. Plus, since sampling can lead to random errors in the total mass, by discounting negative tendencies in the timeseries it could theoretically be possible that you bias the result. In any case, estimating an injected SO2 amount is such a natural part of assuming the exponential model of SO2 decay that it would fit well here and provide the basis of your conclusions about the method.

This is a good point, and we have reworked Section 6 and updated Table 2 to include this estimate of the total SO2 burden. Please see the revised document for the changes.

- In your response to referee comment “Is there any possibility to infer the disturbing seasonal cycle from comparing different years with less volcanic influence?” I don’t understand your reply that “The spread from year-to-year is larger than the intra-annual variability.” It appears from your plot that the intra-annual spread is around 500 Gg, and the spread between years is around 100-200 Gg. It appears that the referee’s idea could be useful and could decrease the uncertainty in the fits, given that the spread in non-volcanic years appears potentially smaller than the spread in the example shown in Fig. 3.

Thank for you bringing this up. Our point could have been stated more clearly in our initial reply. We are trying to emphasize that at each day of the year, there is a spread of around 100-200 Gg. Supposing we take the mean of the background years as the reference seasonal cycle, the actual disturbing seasonal cycle could be $\pm$50-100 Gg offset from the reference. This would impart a large bias on the calculated decay rate, thus limiting our ability to constrain the uncertainty.

The interfering background seasonal cycle is interesting in its own right, and to our knowledge under-explored in the literature. Given that it is likely due to O_3 and HNO_3, perhaps one might be able to calibrate the seasonal cycle based on observed values of these trace gases. However, we feel this would be beyond the scope of the current study. Nonetheless, we have added further discussion on this point (lines 305-312) in Section 4.2 as well as a supplementary figure (S2).

> “The approach outlined above samples possible seasonal cycles using the observed time series in a given year of an eruption. One could also potentially infer the interfering seasonal cycle from years without large volcanic eruptions; however, this presents its own challenges. There is a 100 to 200 Gg spread in SO_2 mass on any given day of the year in non-volcanic conditions (Fig. S2). Using the mean across years for the background seasonal cycle could result in an offset of $\pm$50 to 100 Gg from actual disturbing seasonal cycle. This difference is large enough to impart a significant bias in the estimated decay rate, thus limiting our ability to constrain the uncertainty. The seasonal cycle and inter-seasonal variability in MLS SO_2 is interesting in its own right: it warrants further investigation and could potentially be calibrated based on the observed amounts of O_3 and HNO_3 but this is beyond the scope of the current paper.”