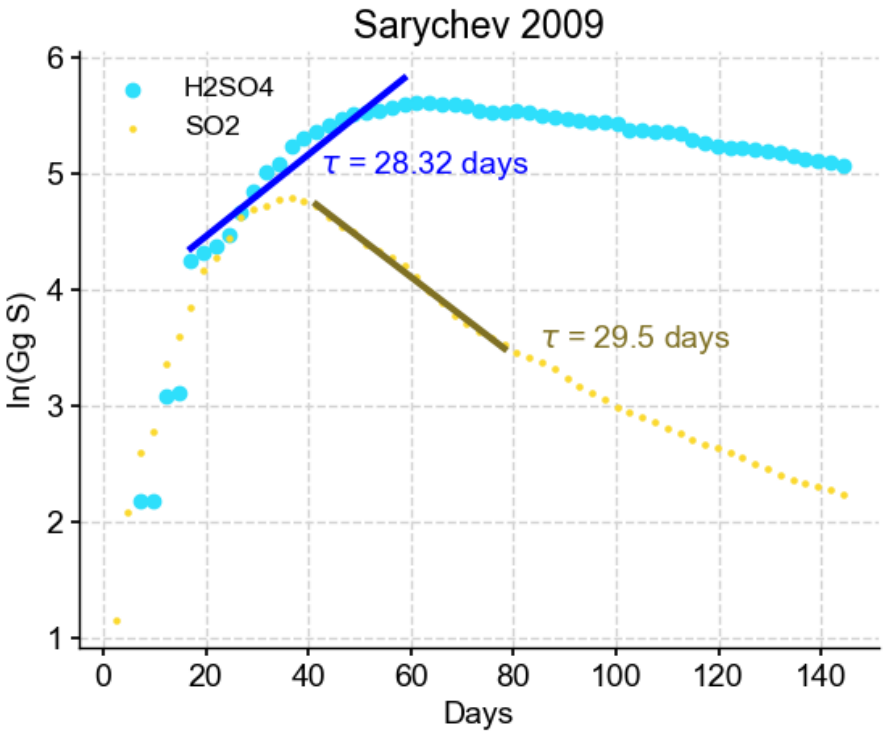
Quantifying the decay rate of volcanic sulfur dioxide in the stratosphere

Response to reviewers in red

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 a discussion about this point to the introduction (lines 49 to 51 in the revised manuscript), as well as a supplementary figure, which is copied below (Fig S1.)



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 140 to 144 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 3.3 (lines 314 to 320 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 22km 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 185 to 188 in the revised manuscript):

> "Given the variation in tropopause height with latitude, the 10 to 14km and 14 to 18km 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).

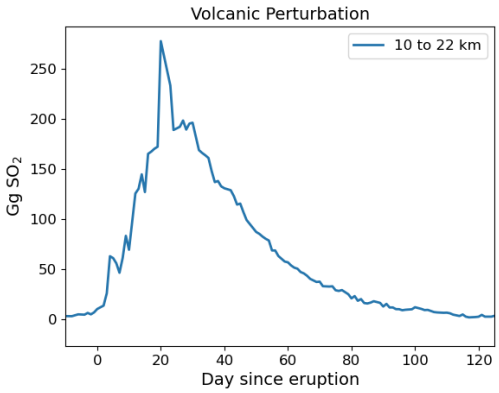
There is evidence in self-lofting of volcanic plumes, similar to that seen in wildfire plumes (e.g., Khaykin et al., 2022; https://doi.org/10.1038/s41598-022-27021-0). However, this has not been noted for the volcanoes analyzed here. Nonetheless, it is a potential source of uncertainty.

Both of these points have been elaborated on in the second paragraph of Section 3.3; see lines 323 to 336 in the revised manuscript.

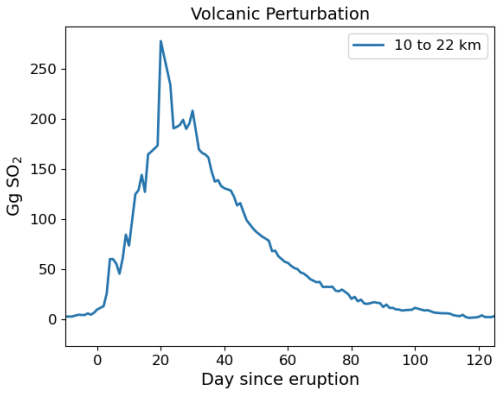
The authors are using a zonal mean SO2 on a 10° latitude grid (ln 153). It seems like they could also construct a tighter latitude grid (say 5°) and a longitude grid and select high SO2 regions which might reduce the uncertainty (e.g. Fig. 2). I would like to see how this affects their decay rate and 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 375 to 379 in the revised manuscript to better emphasize this:

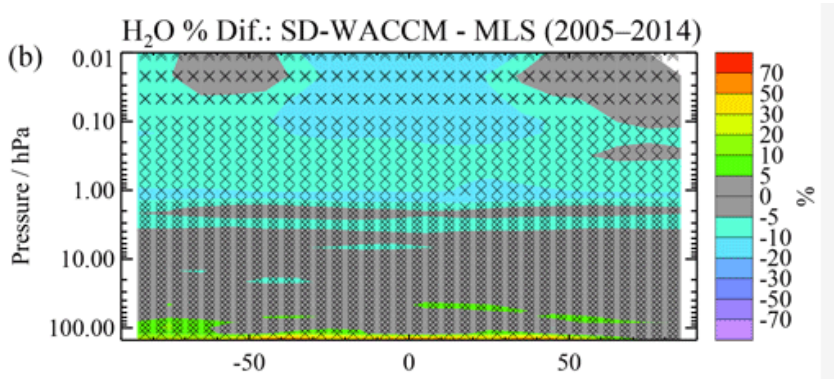
"We compare the results from MLS, MIPAS, and WACCM with SO_2 retrievals from the Ozone Monitoring Instrument (OMI). OMI is a popular choice in recent work examining the decay of SO_2 following eruptions (e.g., Carn et al., 2022; Zhu et al., 2020; Krotkov et al., 2010), and we include an analysis of it 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 4 to highlight this point (lines 393 to 398 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 3.3 (lines 358-360 in the revised manuscript):

> "One reasonable issue 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

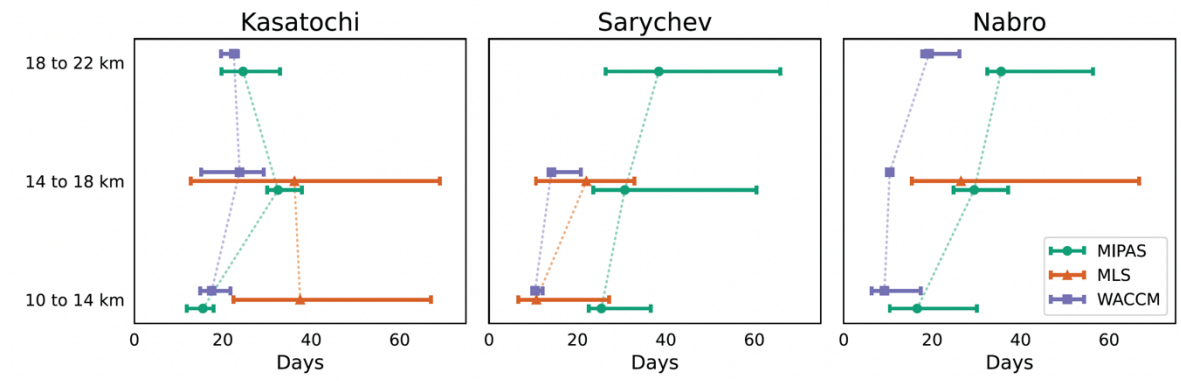
We think the legend sufficiently conveys which lines correspond to the different layers, and adding labels directly on the plot would add unnecessary clutter to the figure.

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 4 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.