Quantifying the decay rate of volcanic sulfur dioxide in the stratosphere

Response to 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.

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 ±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."