

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.

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.
- (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 McKen 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.

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”

Ln 30 ‘once the plume reaches the stratosphere’

Ln 41 There a lot more volcanic aerosols impacting climate references (see Robock, Stenchikov, etc.)

In the literature review, you should cite McKen et al. (1984) here as well as in line 173

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

Ln 107 Livesey is spelled wrong.

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

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

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.

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.

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

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.

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.

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.

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

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.

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

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.

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

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.

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

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