

## ***Interactive comment on “Sulfur dioxide (SO<sub>2</sub>) from MIPAS in the upper troposphere and lower stratosphere 2002–2012” by M. Höpfner et al.***

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We thank referee 2 for detailed comments - especially regarding the differences between derived e-folding lifetimes from limb and nadir sounding instruments. Below we address all comments (bold face) one by one.

**P2, I 76: Please add a reference to the early study of Yang et al, Yang, K., X. Liu, P. K. Bhartia, N. A. Krotkov, S. A. Carn, E. J. Hughes, A. J. Krueger, R. J. D. Spurr, and S. G. C1572 Trahan (2010), Direct retrieval of sulfur dioxide amount and altitude from spaceborne hyperspectral UV measurements: Theory and application,**

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**J. Geophys. Res., 115, D00L09, doi:10.1029/2010JD013982**

We have added this reference.

**Table 2 should be extended for higher SO<sub>2</sub> columns (a maximum of 10 DU is not enough). How this error can influence the lifetime estimates?**

As suggested by the reviewer, we have redone the simulation for a five time enhanced value (500 ppbv) of the maximum SO<sub>2</sub> volume mixing ratio (vmr) reported in the original table (in comparison MLS observed maximum vmr values of about 400 ppbv after the eruption of Kasatochi in 2008). As expected the underestimation of the resulting vmr error increased further to -77% (-54% for the 10-25 km layer column amount). These results are appended to Table 2. We judge the influence of the saturation error on the estimates of SO<sub>2</sub> lifetime to be small. For determination of lifetimes we use data only starting some weeks after the eruptions when observed SO<sub>2</sub> volume mixing ratios are below about 4 ppbv and, thus, well in the region where saturation errors are below 10% for maximum vmr values and less than 1% when considering partial column amounts.

**Section 2.4.1 a short paragraph introducing ACE-FTS would be useful, including references. Please define ACE-FTS and its satellite platform. Please provide general information on instrument characteristics (e.g., spectral resolution).**

The following description of the ACE-FTS instrument has been added to the manuscript:

‘ACE-FTS is one of the instruments belonging to the Atmospheric Chemistry Experiment (ACE) space mission launched in August 2003 (Bernath et al., 2005). The Fourier transform spectrometer measures infrared solar occultation spectra from 750–4400 cm<sup>-1</sup> with a spectral resolution of 0.02 cm<sup>-1</sup> at sunrise and sunset during each orbit. The vertical resolution of the retrieved profiles of atmospheric trace gases is about 3–4 km as set by the instrument’s field-of-view. More specific information on the reconstruction of SO<sub>2</sub> vertical distributions from ACE-FTS measurements can be found

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in Doeringer et al., 2012.'

**Section 2.5.1: please rename 2.6 (there is no section 2.5.2) The significance of this section is limited compared to the other sections. It has the merit to exist but no conclusions can really be drawn notably for P2 because of the remaining bias (Fig 5).**

The chapter has been renamed.

**Page 5, l 419: Please recall what is the typical value for the half-width of the averaging kernel.**

We have repeated the information (3-5 km) at this text passage.

**Page 5, l 460-473: it would be good to assess the importance (in %) of the 3 effects. At least the second effect (saturation) could be evaluated (see previous comment on Table 2). Is the use of alternative spectral windows (less affected by saturation) feasible?**

To realistically disentangle and quantify the three effects would require an extensive study: in a kind of Monte-Carlo simulation one would have to calculate realistic volcanic eruptions including the effect of the inhomogeneity along the limb line-of-sight of MIPAS. In addition to the SO<sub>2</sub> plume, the volcanic aerosol distribution for various scenarios would have to be simulated. This kind of study reaches by far beyond the scope of the actual paper since it also would not help in a straightforward manner to improve the MIPAS observations directly following the volcanic eruption.

One argument why we think that the MIPAS cloud clearing is the most important reason for the differences between MLS and MIPAS directly after strong volcanic eruptions are the maximum vmr-values of SO<sub>2</sub> retrieved from MIPAS spectra: around 13 ppbv. A comparison with Table 2 shows that in case of saturation effects this would amount in about 18 ppbv of SO<sub>2</sub> in reality. Such a relatively small difference cannot explain the large difference between MIPAS and MLS directly after the eruption when MLS

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observes maximum concentrations of around 400 ppbv of SO<sub>2</sub> (e.g. Figs. 1 and 2 in Pumphrey et al., 2015). Since the difference in sampling geometry between MIPAS and MLS is also not so large that it could easily explain the observed differences, the strongest effect is most probably the applied MIPAS cloud/aerosol clearing. We have stated this more clearly in the text.

With regard to the comment on spectral windows: the retrieval has been set up using the spectral region around 1370 cm<sup>-1</sup> which contains the strongest signatures of SO<sub>2</sub> in the spectral range of MIPAS, like in Höpfner et al., 2013. However, in order to reduce saturation effects, for the single retrievals we have added lines from the region around 1130 cm<sup>-1</sup> where the intensity of the SO<sub>2</sub> lines is weaker. This information has been added within the chapter 2.2 where the set-up of the retrieval is described.

**Section 3.2 : My main point of criticism on this study is related to the lifetime estimates.**

**a) P6, l491: it is unclear what is done to appreciate the “linear behavior”. A fitting line could be added on Fig 13 for illustration purpose.**

For illustration, we have added a new Figure 14 (see Fig. 1 below) to the manuscript which shows the MIPAS data of Fig. 13 in logarithmic scale. In this Figure, several fits to the quasi-linearly descending region of the data are shown. The results for the lifetime are presented in the legend. Further we have added a line representing a lifetime of 10 days (bold grey line). From comparing these, it is clear that such a short lifetime is not compatible with the dataset.

**b) The authors retrieve 13.3d, 23.6d and 32.3d which are really high values and are only supported by limb measurements from MLS but not by nadir sensors, except the estimation by Karagulian et al. (2010), which is also questionable (read below). Therefore my question is whether it could be that the limb lifetime estimates are biased high due to a different sampling/coverage of the volcanic plumes than the nadir sensors.**

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It is difficult to explain the longer lifetime which we obtain from the limb-measurements with sampling issues. E.g. from perspective of MIPAS, we generally use the measurements starting some weeks after the eruption, i.e. when the plume has already spread over a larger area. Also we see that our retrieved lifetimes are clearly dependent on the altitude, which is expected in the upper troposphere/lower stratosphere where SO<sub>2</sub> concentrations are controlled by the availability of OH. At lower altitudes, additional removal through clouds should significantly reduce the lifetime. We propose two explanations for the SO<sub>2</sub> lifetime differences between limb and nadir sounders: (1) the sensitivity of nadir viewing instruments to SO<sub>2</sub> is much smaller compared to limb-sounders. When the plume becomes more and more diluted over the globe, the small concentrations get smaller than the nadir-sounders' detection limit. In that way lifetimes are artificially shortened. This effect has already been discussed in Haywood et al. (2010) who derived from IASI observations of the Sarychev eruption a smaller lifetime of SO<sub>2</sub> in comparison to model calculations. (2) Nadir sounders are vertically sensitive down to the middle troposphere where the lifetime of SO<sub>2</sub> is much smaller compared to the UTLS region. This means that different lifetimes from very different altitudes are mixed, also from heights below the lowest point where limb-sounders deliver data.

**c) Clarisse et al. (2012) presented new results for the Kasatochi case using IASI that are improved compared to Karagulian et al., 2010. Therefore the 18d estimation lifetime is probably outdated (read end of section 4.1 in Clarisse et al., 2012). Hence, lines 557-562 (P6) are probably not applicable anymore. Note also that Theys et al. inferred a lifetime of 7 days for the eruption of Puyehue, i.e. far from the estimate of 32d given in Table 3.**

We agree with the reviewer that there is a disagreement between the SO<sub>2</sub> lifetime estimates between the limb-sounders (MIPAS and MLS) and the nadir sounding instruments. We reformulated the discussion on this issue and added the information on lifetimes from Clarisse et al., 2012 and Theys et al., 2013 in Table 3.

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Added text:

'From nadir sounders in case of Ka08, Karagulian et al. (2010) derived a lifetime of 18 d. This value, however, has been challenged by Clarisse et al. (2012) who determined similar values as reported by Krotkov et al. (2010): 8–9 d. For Sa09 Clarisse et al. (2012) showed a comparable time dependence as Haywood et al. (2010) pointing to a lifetime of around 10 days. Thus, there is a clear difference between SO<sub>2</sub> lifetime estimates from nadir and from limb-sounding instruments. Figure 14 illustrates this discrepancy by comparing a decay time of 10 days to the MIPAS observations from Fig.13 in logarithmic representation. Haywood et al. (2010) have noted a similar difference between their nadir sounding observations and results from model runs. These differences have partly been explained by the SO<sub>2</sub> detection limit of the nadir measurements leading to lower lifetime estimates upon dispersion of the plume. A further contribution might also stem from the vertical sensitivity of nadir sounding instruments in combination with vertically varying decay times of SO<sub>2</sub>: nadir sounders also sample air from altitudes lower in the troposphere which are not seen by the limb-instruments and where the lifetime of SO<sub>2</sub> is probably smaller than at higher altitudes.'

**d) It would be interesting to redo the lifetime calculation for the integrated total SO<sub>2</sub> mass (not resolved for the 3 atmospheric layers). As it is now, the estimation of  $\tau_i$  assumes that there is no transport/exchange of SO<sub>2</sub> between the different layers, which is not guaranteed.**

We have performed the proposed calculations for Kasatochi, Sarychev, and Nabro and found a mean lifetime of 16 days. This is inside the range of lifetimes we get for the three altitude regions (13.3 d, 23.6 d, and 32.3 d for 10–14, 14–18, and 18–22 km, respectively) and slightly smaller than the mass-weighted mean lifetime of 19 days. This difference might point to an effect of mixing between the layers, but might also be due to an uncertainty caused by the single errors on mass and lifetime estimates.

**e) Since the mass estimates are not independent of the lifetime which is assumed**

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**(when not fitted from the time series), it would be good to use a lower lifetime value (say 10 d, ie in better agreement with the nadir estimates) and check if it improves the agreement with the nadir data for the total mass.**

Given the fact that such small lifetimes like 10 d are far from being compatible with the limb-observations, as discussed above (see also Fig. 1), we do not think that such an exercise would deliver more insight.

**P 7, 1595-599: I think this is a quite unlikely explanation. The typical time scale for the atmospheric circulation is much larger than the SO2 lifetime.**

We do not agree with the referee in this aspect. As has been shown in Höpfner et al., 2013, there is downwelling of high values of SO<sub>2</sub> during polar night where its lifetime is much enhanced due to lack of OH. The conversion into H<sub>2</sub>SO<sub>4</sub> aerosols starts during spring when sunlight and OH becomes available. This leads to the so-call 'Condensation Nuclei (CN)-explosion'.

**Fig. 10: it might be better to use a non-linear color scale to better represent the range of values**

We have decided to leave the plots with the linear colour scale in the paper. In addition, to be able to view the whole range of SO<sub>2</sub> values, a supplement containing Figures 10-12 represented with a logarithmic colour scale will be added.

**P1, I 52 : SO2 should be in parenthesis (as a definition)**

Done.

**P 7, I650: 'depolarisation'-> 'depolarisation'**

Was already corrected in the published ACPD version of the manuscript.

## References

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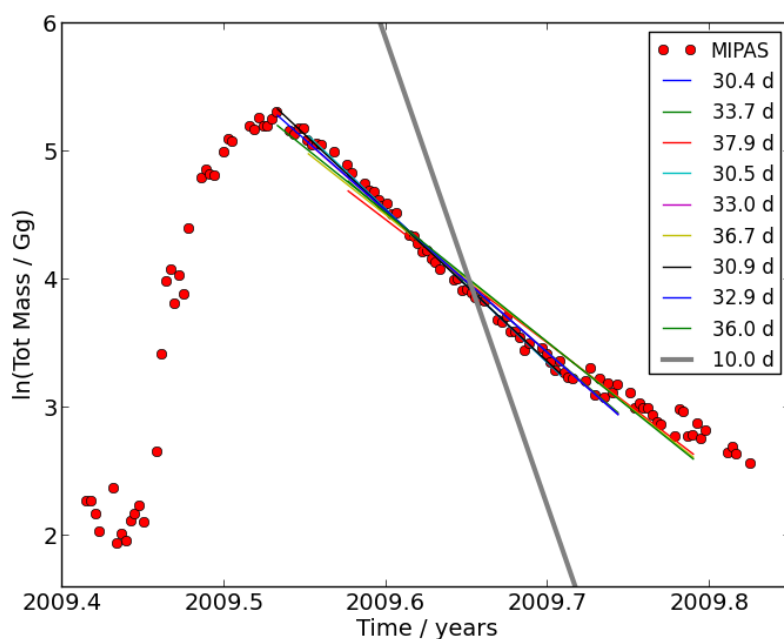
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Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/15/C2827/2015/acpd-15-C2827-2015-supplement.pdf>

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 15, 5801, 2015.

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**Fig. 1.** Logarithmic representation of total mass of SO<sub>2</sub> above 146.8 hPa from MIPAS (red dots) from Fig. 13 in comparison with fitted lifetimes (thin solid lines) and a lifetime of 10 days (bold grey).

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