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Atmospheric Measurement Techniques Discussions

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Interactive Comment

Interactive comment on "Observations of volcanic SO_2 from MLS on Aura" *by* H. C. Pumphrey et al.

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Referee #1

No comments were received from referee 1 in the open discussion stage.

Referee #2 General comments

A general point which I miss, is a discussion of the estimated errors of the MLS SO_2 data and a comparison with the validation results. This might indicate whether there are other error sources which have not been taken into consideration for estimating errors. This would be valuable information for data users to decide whether estimated errors from the data document could be used.

I have looked into this and note that the estimated errors supplied with the data are

2-4 times smaller than the standard deviation of the data in regions where there is no volcanic SO_2. We have added text to this effect to the paper.

Specific corrections

P7884L13 [in the abstract] 'The agreement is good' Could you state this more quantitatively? (e.g. within . . . % . . .)

The agreement referred to here is between MLS and OMI total columns. We have gone for "within 1 DU" rather than "within X %" as it is not clear that the differences between the two instruments are proportional to the total column.

P7885L1 'A volcanic eruption is the only such process that is of any importance.' This statement is very strong. However, what about heavy fire events (pyroconvection)? Also there are discussions about how much water vapour reaches the stratosphere via strong convection. The Asian monsoon circulation is a further mechanism to transport pollution relatively fast to high altitudes. These processes should be mentioned and references given.

What we meant was that a volcano is the only process that has been observed to loft SO_2 to that altitude. Pyroconvection certainly can loft a variety of biomass-burning products to that altitude (Pumphrey et al., 2011) but we are not aware of a case in which those products have been observed to include SO_2. We have edited that sentence to make it less sweeping and more specific. The preceding sentence makes it clear that the offending sentence was only referring to processes at the surface and not to processes such as the Asian monsoon, which the volcanic products might encounter after being emitted. Convection processes elsewhere in the atmosphere are unlikely to transfer SO_2 to the stratosphere because it is too soluble in water.

P7885L15 : There are also papers describing height information retrieval from IR-nadir sounders: e.g. Carboni et al., Atmos. Chem. Phys., 12, 11417–11434 and Clarisse et al., Atmos. Chem. Phys., 14, 3095–3111, 2014.

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The paper as discussed already references Clarisse et al. (2014). We have added a reference to Carboni et al.; apologies to the authors for missing it out.

P7888L7: In Fig. 4, the 46.4 hPa data seem to be most heavily influenced by the seasonally varying bias problem. Is this the feature which is described here? However in Fig.11 lowest values of total mass in the background seem to be reached by mid of 2008 while the vmr values at 46.4 hPa seem to have a maximum in Aug/Sep?

Fig. 11 shows an estimate of the total mass in a given latitude band; this will be affected by all pressure levels used. The higher pressures dominate this calculation, so the seasonal cycle at 46 hPa will have a much smaller effect than that at 215 hPa. Fig. 4 shows mixing ratios in each pressure as a separate line. Furthermore, when there is no plume it shows the three highest values during the day. If there is no plume, then the highest values of SO_2 occur in the southern polar vortex. As already discussed in the paper, the apparent seasonal cycle in SO_2 as observed by MLS is thought to be caused by interference from O_3 and HNO_3. Both of these species have very different values in the vortex than out of it.

P7891L19 'Both the latitude range and the highest pressure used in the vertical integration are chosen separately for each eruption.' What have been the criteria for the selection of the latitude range and the pressure?

Both the latitude range and the highest pressure used in the vertical integration are chosen separately for each eruption in order to encompass all of the profiles and levels showing enhanced SO_2 from that eruption. We have added the previous sentence to the paper to clarify this point.

P7894L4: [and hence Fig. 13] Can the SO2-background offset of the 240 GHz channel be identified in all three channels? How different is it between channels?

An offset which depends on time and latitude is present in all three bands. It is different in all three, adding weight to the hypothesis that it is not due to SO_2 and is due to interference from other species. In all cases its magnitude is smaller than 10 ppbv and usually in the range 2-5 ppbv. The correlation shown in Fig. 13 is not affected by the background offset. In the absence of volcanically-enhanced SO_2 there is little or no correlation between the different SO_2 products; a scatter plot like Fig. 15 consists of an elliptical cloud of points with a radius of about 10 ppbv centred less than 5 ppbv from the origin.

P7896L10 'The agreement between MLS and OMI is good': Could these results be discussed a bit more quantitatively? What are the biases (standard-deviations) of the difference between the two instruments? Do these differences fall within estimated errors of the MLS SO_2 product?

As noted elsewhere in this response the estimated errors in the MLS SO_2 product are smaller than the standard deviations in the data in regions with no volcanic SO_2. The best way to estimate the random error in the MLS total column is to calculate its standard deviation in the un-enhanced regions. For Manam (above 147 hPa) this is about 0.36 DU. For Montserrat and Manam (above 100 hPa) it is about 0.25 DU. For Kasatochi (above 215 hPa) the error estimated by this method is 0.85 DU. In all cases the random error estimated in the absence of SO_2 is small compared to the variability when SO_2 is enhanced. This implies that the main cause of scatter in the MLS-OMI comparison is the large spatial variability of the volcanic SO_2. We have added some text to the paper to this effect.

Technical:

P7919, Fig.15, caption 'This is really more like an estimate of the column above 121 hPa': Such a discussion should better appear in the text rather than in the Figure caption.

We have moved that discussion into the running text.

P7901L11: 'Version 2.2 Level 2 data quality and description document': Should this

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not read: 'Version 2.2 and 2.3 Level 2 data quality and description document' ?

It should, yes. The name of the document has been changed since its first release to reflect the fact that the version 2 software received a significant change and the version number was changed to 2.3x instead of merely from 2.2x to 2.2y. We have updated the reference.

Referee #3

General comments The referee suggests that

Table 1 lists a lot more eruptions than most other references which is useful for the modeling community. Figures 3 and 4 suggest that this can be even expanded, for example by eruptions early in 2007 and 2011 which are 'observed' several times.

We have tried to include every eruption from which SO_2 is observed by MLS in table 1.

In early 2007, figures 3 and 4 show Piton de la Fournaise and Jebel-at-Tair. Both of these eruptions are listed in table 1. In 2011, the only clear eruptions are Grimsvötn, Puyehue-Cordón Caulle, and Nabro. The data have a couple of other "spikes"; closer inspection suggests that these are artefacts in the data and are not associated with a volcanic eruption. The paper as discussed already notes this (Page 7889 line 11).

Specific corrections

Page 7887, line 22: I.e. the whole latitude range? Yes, that is the whole latitude range. We have added an explicit statement to that effect in the caption to figure 1.

Page 7888, line 4: Say explicitly that [the 7.7 standard deviation cutoff] is 2 to 3 orders of magnitude above background.

This depends on what you mean by "background". The 7.7σ cutoff is typically in the 50 ppbv – 100 ppbv range. This means that it is perhaps 20-50 times greater than the sort of daily averages reported by MLS (which, as we explain in the paper are spurious). The background values reported by Höpfner et al. (2013) are in the range

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5 pptv to 20 pptv, so the the 7.7σ cutoff is about 2 orders of magnitude above this. We have added a sentence to the paper to this effect.

Page 7888, line 7f: Here the latitude and time dependent bias should be mentioned explicitly. Done.

Page 7889, section 3: Why the daily maxima are selected in Fig. 4? Please expand, or better, correct the data by the seasonal bias in the figure and show the maxima only if they are over the threshold for Fig. 3. Fig. 4 like it is suggests that there are volcanic (or anthropogenic) events available all the time raising maximum SO_2 almost 2 orders of magnitude above the background. If the authors think that this is the case this should be said in the text. Also in Fig. 4 the data for 316 and 46 hPa might be skipped.

Figure 4 was added at the pre-discussion stage, at the request of referee #3, who asked us to show the measured mixing ratios in the volcanic plumes in Figure 3. The main difficulty with it is that its purpose was to show the mixing ratio in the plumes. But you then have to think of what you want to show at the times when there is no plume. The choice to show the maximum values observed that day introduces the least amount of arbitrary decision-making to the plot. We have considered a couple of alternatives and decided that they are less satisfactory than the plot as it stands. One possibility is to show no data at all where a plume has not been detected. This would make the plot look bitty and would make it hard for the eye to see the smaller events. Another idea would be to show the daily mean or median in the periods where there was no volcanic SO_2. This would make the volcanic periods look more different from the rest of the time series than they really are. We think that the best way to address the reviewer's concern is to make the figure caption a little more explicit, so this is what we have done.

Page 7890, line 25: There was also an eruption of Soufrière Hills in 2010. So there was. In fact, we even list it in table 1. We have changed the end date to 2010. We have also added a reference (Wadge et al., 2014) to the special edition of the Geological

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Society Memoirs devoted to the recent eruptions of this volcano.

Page 7891, line 14ff: Seasonal bias mentioned late. The correction function should be given somewhere (Appendix or supplement). We have now mentioned it earlier as noted above. We have not constructed a correction function for the entire dataset and are not certain that it would be useful or practical to do so. As the paper already states, the correction for the purposes of calculating the SO_2 burden was done by fitting annual and semi-annual cycles. That these cycles were sinusoids was implicit: we have added a word to make it explicit.

Page 7892, line 1: Point to $\log_e$ *axis in Fig. 12.* We are not sure how the referee wants this pointing to be done. We have added a sentence to the figure caption to make it more explicit.

Page 7894, line 18: The MLS column should be between the STL and TRM OMI products.

Where the SO_2 is mostly at 100 hPa (~ 16 km) or 68 hPa (~ 18.7 km), the comparison with the STL product is appropriate — this covers the Manam and Montserrat events. Furthermore, the OMI SO_2 README file states "STL data are intended for use with explosive volcanic eruptions where the cloud is placed in the upper troposphere or stratosphere (UTLS). At these altitudes the averaging kernel is weakly dependent on altitude, so that differences in actual cloud height from $\sim$17 km produce only small errors". We have added sentence to the paper to this effect and have not included a comparison against the TRM product in the paper, even for the Kasatochi event.

Nevertheless, it is worth noting briefly that for the OMI SO_2 data points compared to MLS for Manam, Montserrat and Rabaul the TRM product is very close to $1.25\times$ the STL product. The STL product agrees well with MLS in these cases, to the extent that the TRM product is clearly larger than the MLS values.

For the Kasatochi event the situation is less clear. Where there is a large amount of

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SO_2 at 215 hPa or below, the STL and TRM products are similar in size, or the STL product may even be slightly larger.

Page 7895, line 6: For which units (SI?) are the equation and the conversion factor? Is f volume or mass mixing ratio? The notation in the brackets is very special and requires more explanation since the given reference is difficult to access (link available?).

We have fixed the reference to add a link to the document referenced (and, in the process, fixed a spelling error in the reference noted by the reviewer in his technical corrections). The formula is in SI units (apart from the mole mass being in grams on account of the definition of the mole and the Avogadro constant) so the answer comes out in number of molecules per m^2. The mixing ratio, f, is a volume mixing ratio. We have expanded the description to clarify which units are being used.

Page 7897, section 6.3.3: This should be expanded.

The IASI SO_2 data are not, to my knowledge, available to the scientific public. There is little we would be able to do here other than paraphrase the referenced papers. An interested reader would be better served by following up the references for himself.

Table 2: For the use with model simulations and better comparison with the other estimates it would be good to provide additionally the values for 147 hPa and 100 hPa for all volcanoes where it is available.

Which values, though? The highest value observed? The average (in some sense) for the plume (defined in some sense)? We do not think that we can easily add another column to Table 2 which will enhance its value. It should be remembered that the MLS data are easily available and the format is well-documented; any serious attempt to compare the data to a model or to another type of measurement would probably need to be done with the actual data. One purpose of publishing the current paper is to encourage the use of the MLS data in this kind of study.

Technical corrections

Page 7898, line 21: This should be ppbv or is line 10 on page 7888 wrong?

This should indeed be ppbv. Fixed.

Page 7901, line 1: Typo. Fixed — this was in the references as noted above.

Page 7904, caption, line 4: Something missing. Last column of table: ',' missing between references (2 times). Fixed.

Figure 13: Better use legend to explain colors. Refer at least to Table 1. We added a line to the text noting what the colours were for and added a reference to table 1 to the figure caption.

Figure 15: Remove '+' symbols. What is ""?* We have removed the "+" symbols. There are no "*" symbols. There are some $\times$ symbols with a vertical error bar through them — and in one case that error bar is a similar size to the $\times$ symbol.

Figure 16: I don't see data for 68.1 hPa There is some, although it is mostly buried in the cloud of points around the origin. We have not removed it from the legend, partly to make the point that the points with enhanced SO_2 all have their peak at a lower altitude than the 68 hPa level.

Other changes made by authors

- Small corrections to references, including changing Höpfner et al. (2013) from Atmos. Chem. Phys. Discuss to Atmos. Chem. Phys., as the final version of that paper is now available.
- Extra legend added to Fig. 2 to make it easier to understand.
- Subsection headings for **Major events in more detail** now all include both a name and a date in a consistent manner.
- Figures 3 and 4 brought up to date. (Nothing of interest has happened since

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Kelut in February 2014, but the reader may as well be able to see that this is the case.)

References

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