

Response to *anonymous referee #2*

Anonymous Referee #2

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Zhang et al. provide a comparison of the current operational NASA principal component analysis (PCA) SO₂ retrieval results for the OMI and OMPS satellite instruments, focusing on three global regions affected by anthropogenic pollution during 2012-2015. Based on daily and annually averaged data, the results indicate that the currently operating (and upcoming) OMPS instrument(s) allow continuing the long-term global SO₂ pollution monitoring by OMI.

The paper is well written. However, besides considering the following specific comments, I think the authors should put a little more effort into analyzing the remaining differences between both satellite instruments (e.g. spatial resolution vs. daily coverage) before final publication in AMT. While the comparison of the OMPS and OMI SO₂ results shows a general agreement between both instruments (which is good to see, but not surprising), not much information is given about the discrepancies and how good both datasets really agree if they would be used for a quantitative long-term monitoring of anthropogenic emissions. Especially for the SO₂ emissions over China it would have been nice to see a more detailed case study, e.g. including a plot with monthly total SO₂ masses for OMI 2004-2015 directly compared to OMPS 2012-2015.

We thank the referee for the detailed review and have made changes to the revised manuscript following these suggestions that have improved the paper. Please see below our responses to the main suggestion and specific comments.

As the referee suggested, we derived SO₂ emissions from both OMI and OMPS. Four years (2012 -2015) of OMI-OMPS SO₂ emissions are compared in a new section in our manuscript. Two new co-authors contributed to this section.

Specific comments:

Abstract, l.18-22: At this point of the paper it remains unclear what the authors mean by the terms 'temporal' and 'spatial' correlation. It would be helpful to clearly state somewhere in the first sections of the manuscript that a linear regression fit is applied to the daily SO₂ masses for an extended time period (with the results here called 'temporal' correlation) and to the single grid pixels of yearly averaged SO₂ column (results here called 'spatial' correlation).

A: We have added explanations of the terms 'temporal' and 'spatial' correlations in the beginning of Section 3 as suggested.

Abstract, l. 22-23: The choice of $r > 0.6$ (and only focusing on the Mexican region) appears a little bit arbitrary in order to demonstrate how 'good' both satellite

instruments agree. Why did you use this specific value? As a first time reader I would have the impression that the numbers for $r > 0.7$ (or higher) are just too low to mention in the abstract. The results presented in Fig. 6 further indicate that only roughly half of the measurements have a correlation coefficient $r > 0.6$ for eastern China and the South African region, which is surprisingly low at a first glance for two instruments evaluated by using the same algorithm and with only some minutes temporal difference. Maybe the main conclusion here is that there are substantial differences for the two satellite instruments if the SO_2 columns are compared pixel by pixel on a daily basis (probably because of the different spatial resolution) and this should be discussed in more detail in the manuscript.

A: We agree with the referee and replaced the sentence in the abstract as “The OMI-OMPS SO_2 mass differences on a pixel by pixel (daily) basis in each region show substantial differences. The two instruments have a spatial correlation coefficient of 0.7 or better on $< \sim 50\%$ of the days.” We also added discussion about this discrepancy in Section 3.2 (Fig. 6).

p. 2, l. 14-17: Could you be a little more specific on that? Isn't it self-explanatory that the main limiting factor in detecting point sources is spatial resolution?

A: “This is because the coarse spatial resolution will dilute the derived SO_2 columns or masses for point sources as compared with fine spatial resolution. This causes additional measurement differences in SO_2 loading from different instruments.” We added this explanation to the revised manuscript. The new emission section also suggests that OMPS coarse spatial resolution is the main limiting factor in detecting SO_2 emissions.

p. 4, l. 15-16: What is the reason for not applying an additional empirical background correction over the Pacific? Figure 1 indicates that there are small (but noticeable) systematic differences for the two satellite instruments.

A: We are not applying an additional empirical background correction over the Pacific because the remaining biases are relatively small (compared with the previous algorithm) and may be reduced when we apply planned improvements in the AMF calculation. In the future, we will explore the use of an empirical correction. The small systematic differences between two satellite instruments are mainly due to differences in the AMF factors. We added statements regarding future work in the revised manuscript.

p. 4, l. 26-27: It is difficult to distinguish the volcanic SO_2 emissions from Popocatépetl from the anthropogenic emissions in the Mexico City area. Based on daily regional OMI SO_2 column maps (e.g. available via <http://so2.gsfc.nasa.gov/>) and because of the increased sensitivity of the satellite instruments to SO_2 at higher altitude, it is quite likely that the main emissions in the considered region (or at least a large fraction) are of volcanic origin. It's however interesting to see that the results agree best over Mexico compared to the other two scenarios (again probably because of the increased sensitivity). Certainly, it would have been better to focus on another region with strictly anthropogenic emissions instead of Mexico (e.g. the copper smelters in Norilsk,

Russia), especially because there is a similar (more comprehensive) AMTD paper by the authors that strictly focuses on volcanic SO₂ emissions. These problems should be discussed in more detail in the revised version.

A: We chose these three regions to compare and contrast different locations in the world, including both hemispheres and the tropics at different latitudes. We agree that the Mexico region is influenced by Popocatépetl eruptions. Daily SO₂ mass correlation in the Mexico region is the highest. It could be due to volcanic eruptions. We added this in the revised manuscript.

Figure 2: Unfortunately, the colorbars are all restricted to ± 2 and ± 0.5 DU. Please scale to maximum/minimum values for each region. Otherwise the main enhancements are always shown in red/white and it remains unclear how large the differences really are. Please indicate that the lower figures show "OMPS - OMI".

The most remarkable discrepancies appear for the highest columns. What are possible reasons? For the Mexican region, I think this is another indication that the main part of the enhanced SO₂ columns is caused by volcanic emissions. The location of the volcano as well as the city centre should be marked in Figure 2.

A: We rescaled the color bar based on your suggestions. The locations of the city center and volcano near the SO₂ peak are marked on the figures. We did add 'diff = OMPS-OMI' for the lower figures in the figure caption.

p. 4, l. 32: As already mentioned in the previous comment: The OMPS SO₂ columns are only lower than those from OMI for the highest OMPS/OMI values for Mexico/South Africa. For large parts of Mexico, the OMPS columns appear higher. Please discuss possible reasons.

A: We discussed possible reasons in the revised manuscript. They include spatial resolution, boundary layer profile assumptions, and time differences for emissions that are rapidly transported.

p. 5, l. 1: What data are included in the 'annual regional averaged SO₂ columns'? Is it the data from the area shown in Figure 2? If so, please name coordinates of the considered areas.

A: Yes. Annual regional averaged SO₂ columns data are from the area shown in figure 2. We added coordinates of those areas in the text.

p. 5, l. 2ff: Please explain the terms 'spatial/temporal' correlation.

A: We added the explanations of two terms in the beginning of Section 3.

p. 5, l. 2: Please give the exact number (not just '>0.9') as you did for the following 2 cases. The value for China is even the highest in 2012 with 0.96.

A: We replaced '> 0.9' by the specific numbers in each region.

p. 5, l. 3: The patterns appear to be all similar, even for lower VCDs. However, it is difficult to compare the data only by taking a look at the given maps, as the maximum

values are all in red because of the chosen colorbar. Please scale to maximum/minimum values for all 3 regions.

A: We rescaled the color bar for all 3 regions. Please see details in the revised figure.

p. 5, l. 4ff: How large is the variation of the values if you include a wider area (how smooth is the background signal)? It would be good to have a reference value over an unpolluted area nearby, as no empirical offset correction is applied.

A: We did a test for a wider area (+1 degree) over Mexico and found the variation of the averaged SO₂ loading reduced by 10% and that correlation coefficients are the same.

p. 5, l. 9-11: Especially in the case of Mexico, the peak OMI SO₂ columns are much higher than those from OMPS. Probably this is the case because of Popocatépetl as the dominating (point) source in the region.

A: We agree that Popocatépetl is a dominant SO₂ source in the region, especially in the peak region. We added discussion in the manuscript.

p. 5, l. 14-15: The given numbers indicate that you should definitely adjust the minimum/maximum of the colorbars in Figure 2: Given a maximum SO₂ difference of -2 DU, probably all Mexican negative values are 3-4 times larger than your minimum colorbar value of -0.5 DU. As your upper limit for the colorbar is +2 DU for the averaged OMI/OMPS data maps, it would be even possible that some of the clearly enhanced measurements differ by a factor of 2. How large are the positive differences?

A: We adjusted the minimum and maximum of color bars in Figure 2. For consistency, we adopted ± 0.7 DU for the difference plots of SO₂ in each region. In the Mexico region, there are only 3 points that are larger than 0.7 DU. They are 1.01, 0.85, and 0.83, shown as the red color in Figure 2. There are 5 points are less than -0.7. They are -1.33, -1.29, -1.36, -1.16, and -2.0. They are shown in white in the figure. Except for the maximum absolute differences (-2.0 and 61%), all those grid boxes with values outside of ± 0.7 DU have less than 50% differences between OMI and OMPS. We discuss possible reasons for those differences between OMI and OMPS in the manuscript.

p. 5, l. 22-23: It would be very helpful to show a total SO₂ mass timeline 2012-2015 for all 3 regions in an additional figure as well as maps for the other years (2013-2015).

A: We added figures that show timelines of 2012-2015 SO₂ masses for all 3 regions.

p. 5, l. 26: The difference in sampling time should be less than 30 minutes, right (typically about 10 minutes)? Do we really expect here large differences due to transportation?

A: We revised our manuscript here. The rapid transport of SO₂ plumes could be one possible reason. Others, such as the boundary layer profile assumption and spatial resolution differences could be additional reasons.

p. 5, l. 30-31: It would be better to give the total (regional integrated) SO₂ mass instead of the averaged SO₂ column.

A: We keep the averaged SO₂ column loadings here for comparison with estimates from Krotkov et al., 2016. The averaged SO₂ masses are given in Table 5. We added some discussion of emissions calculated from the two instruments in Section 4.

p. 6, l. 1: How significant is the change of the total SO₂ mass if you change the area considered for the calculations (e.g. $\pm 1^\circ$ lat/long)? Some emissions over China are certainly left out because of the currently selected area.

A: We did tests with eastern China with current domain + 1° lat/lon; r and slopes have no significant changes. They are increased by only $\sim 0.1(r)$ and $\sim 0.2(\text{slopes})$ for the larger domain. We did not include this in the manuscript.

p. 6, l. 31-33: I don't really get the argument. Do you think that the correlation decreases because the remaining SO₂ columns are close to the detection limit and therefore scatter around a remaining offset? Otherwise I don't see why the decrease of the averaged SO₂ column should lead to more significant discrepancies. Please reformulate.

A: We reformulate the sentence.

p. 6, l. 17: This is only the case for 2014/2015. For 2012/2013 the slope decreases while the r value remains quite stable...

A: We revised this in the manuscript.

p. 6, l. 18-19: What are the total SO₂ masses per year for the individual regions? In my opinion, it would make more sense to give the total emissions instead of the averaged daily SO₂ mass for each year.

A: We added a new part to compare SO₂ emissions from the two instruments. Please see Section 4 in the revised manuscript.

p. 6, l. 21-23: Same argument as in p. 6, l. 31-33: Please make this point more clear.

A: We did this. Thanks for pointing it out.

p. 7, l. 3: Which days were excluded? All days in April/May or just the ones exceeding a certain SO₂ threshold over South Africa?

A: We removed 3 days when SO₂ masses exceed 30 kt and added it to our revised manuscript.

p. 7, l. 16-17: Why did you decide to show only data for August in each year? Including all individual months would further indicate if there are some seasonal differences between OMPS and OMI.

A: August is the month least influenced by SO₂ transported from polluted regions. We did analyses in other months; please see plots in the supplementary material.

Minor comments:

p. 2, l. 4-5 and 10-11: Please add Theys et al. (2015). This is one of the most recent and comprehensive papers about an OMI SO₂ retrieval.

Theys, N., I. De Smedt, J. van Gent, T. Danckaert, T. Wang, F. Hendrick, T. Stavrou, S. Bauduin, L. Clarisse, C. Li, N. Krotkov, H. Yu, H. Brenot, and M. Van Roozendaal (2015), Sulfur dioxide vertical column DOAS retrievals from the Ozone Monitoring Instrument: Global observations and comparison to ground-based and satellite data. J. Geophys. Res. Atmos., 120, 2470–2491. doi: 10.1002/2014JD022657.

A: We added this reference.