

Point-by-point clarification to Referee #1

In the present response letter, we summarize our modifications in the revised version of the manuscript and we provide point-by-point clarifications (in blue color) to the referee's comments and suggestions (in black color). We found the referee's comments very useful and in the right direction in order to improve the scientific quality of the paper. All of the referee's comments were taken into account in the revised version and we believe that it is now much better.

The authors present a trend analyses of ground-level SO₂ concentrations from OMI measurements over the US. This paper is interesting and has high potential, but, in my opinion, currently lacks sufficient contest and motivation. The flow of the paper is good and logical, although some sections are a bit too compact to my liking. I therefore suggest the paper to be accepted after major revisions, considering the text below.

We thank the referee for recommending the manuscript for publication. We have incorporated all the comments/suggestion in the revised manuscript as suggested by referee.

Introduction.

The introduction clearly states the benefits of studying SO₂ from satellite, mentioning its role in the formation of sulfate aerosol and the effect of the latter on climate and environmental and health issues. Related previous work is adequately cited. However, the cited paper of Krotkov et al. [2016] already gives a trend analyses of OMI total column, over the same time period as the current paper, and furthermore indicates (for polluted areas) the close relationship between derived total columns and emissions. Although the current paper studies surface concentration rather than total column, I would like to see a more elaborate text, motivating why studying total column is not sufficient a proxy for emission trend analysis and the connected assessment of health risks. Only one short sentence is currently dedicated to the novel aspects of the paper and at first glance the overlap with previous work seems high. Please expand.

We agree with referee's view point and included more detail on novel aspects of the paper in the revised manuscript. The total column SO₂ is a sufficient proxy for emission trend analysis but is of low utility for the assessment of health risks. We include the sentence "In contrast to total column SO₂, long-term records of ground-level SO₂ concentrations from satellite observations will be directly useful to assess air quality and associated health risks." in the revised manuscript at Page number 2, Line number 18-20.

Section 2. 2.1 OMI: Concise paragraph. Line 3: 'Also, SO₂ variability...' I presume background SO₂ is meant here? Line 6: Please explain the use of the respective thresholds of 0.2 and 65 degree or give an reference.

We have modified the sentence as "Also, SO₂ retrieval variability is reduced by a factor of two in the PCA algorithm relative to the BRD algorithm (Li et al., 2013)" at Page number 3, Line number 8-9, and provided reference for respective thresholds at Page number 3, Line number 18 in the revised manuscript.

2.3: Model information Line 24: It would be good to have a quantitative indication of the thickness of the lowest model layer, so reader not familiar with GEM-MACH can develop a feeling for what is assumed as 'surface concentration'. Along the same line, an indication of the partial column of the lowest layer with respect to the total boundary layer column is missing. The reader is referred to McLinden et al papers for AMF related information, but I think it should be discussed to some extent in the text (here or in the next paragraph).

We have modified the sentence as “The lowest model layer, which is 20 m thick, is taken as ground-level concentration.” in the revised manuscript at Page number 4, Line number 11-12.

We have included the following paragraph on AMF related information in section 2.1 at Page number 3, Line number 9-16:-

“Even though the PCA algorithm directly estimates SO₂ vertical column density in one step using SO₂ Jacobians, the air mass factor (AMF) is effectively fixed at 0.36 (representing summertime conditions in the eastern USA), similar to the BRD algorithm. A better estimation of AMFs is needed for different regions to reduce these systematic errors that result from conditions that do not match these. For this, we re-calculated the AMFs using SO₂ profile information from the high resolution (15 km x 15 km) GEM-MACH air quality forecast model (discussed in section 2.3), monthly-varying surface reflectivity from the MODIS satellite instruments, and an improved identification of snow. More details on Environment Canada Air Mass Factors calculation for SO₂ are discussed in McLinden et al., 2014; 2016b.”

2.4 Estimation of ground-level SO₂ from OMI. I have the same problem with this paragraph as with the previous. A simple connection between observed and model concentration and column properties is given, adopted from literature, but no discussion is given. This would be ok in the case of an extended section 2.3 Line 2: The cited Lamsal [2008] paper on NO₂ is missing from the list of references.

We have included Lamsal [2008] reference and more detail on AMF calculation in section 2.1 in the revised manuscript.

“Lamsal, L. N., Martin, R. V., van Donkelaar, A., Steinbacher, M., Celarier, E. A., Bucsela, E., Dunlea, E. J., and Pinto, J. P.: Ground-level nitrogen dioxide concentrations inferred from the satellite-borne Ozone Monitoring Instrument, *J. Geophys. Res.*, 113, D16308, doi:10.1029/2007JD009235, 2008.”

3 Results and Discussion. The actual results look sound and well described. Line 26/27: Lee et al [2011] paper is missing from the list of references. Also, this paper already derived SO₂ surface mixing ratios from OMI and compared them to in-site measurements, be it only for 2006. Also this motivates a clearer description of the novel aspects of your paper in the introduction.

We have included Lee et al [2011] reference in the revised manuscript.

“Lee, C., Martin, R. V., van Donkelaar, A., Lee, H., Dickerson, R. R., Hains, J. C., Krotkov, N., Richter, A., Vinnikov, K., and Schwab, J. J.: SO₂ emissions and lifetimes: Estimates from

inverse modeling using in situ and global, space-based (SCIAMACHY and OMI) observations, *J. Geophys. Res.*, 116, D06304, doi:10.1029/2010JD014758, 2011.”

We have included the following sentence in the introduction at Page number 2, Line number 16-17.

“In previous studies (Lee et al., 2011; Nowlan et al., 2011), ground-level SO₂ concentrations were estimated for only a one year period using satellite observations over North America.”