

Interactive comment on “Aura OMI observations of regional SO₂ and NO₂ pollution changes from 2005 to 2014” by N. A. Krotkov et al.

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Referee # 2.

The paper deals with changes in NO₂ and SO₂ levels in several regions using satellite-based observations. It includes accurate reference to different emission sources and processes corresponding to different industrial and other human-related activities. The authors present also interesting information on the ratio between SO₂ and NO₂ content with interesting discussion about the changes in political and economic conditions. The paper is well written and the methodology appropriate. I recommend publication on ACP after addressing the following minor comments.

1) Section 2.3 You mention you select only clear sky conditions. Could you comment

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if/how this could affect your results?

- Given the very small threshold for cloud fraction, the shielding effect from the remaining clouds (above the boundary layer) is expected to be very small. We have added new references (McLinden et al., 2014) to quantify remaining cloud related error is less than 20%. We also added discussion about a “clear-sky” bias due to our sampling of OMI data. Indeed, by selecting only clear sky conditions, our sampling of the dataset may have a clear-sky bias due to changing photochemical and weather conditions. Analysis of surface SO₂ and NO₂ concentrations reveals systematic differences under clear-sky and overcast conditions (K. Vinnikov, personal communication, (Geddes et al., 2012)). While the impact of a “clear-sky” bias might be determined from observations of surface concentrations, altitude profiles from which column content can be determined are far fewer. Model study of the differences in NO₂ and SO₂ VCDs under clear versus all-sky conditions shows both negative (for NO₂ from -5% to -50%) and positive (for SO₂ from +5% to +25%) biases (McLinden et al., 2014). Negative NO₂ bias is consistent with a shift in the NO_x (NO₂ + NO) partitioning to favor NO as a result of increased photolysis under clear-sky conditions. Positive SO₂ bias is due to accelerated SO₂ conversion to sulfate in presence of clouds (by aqueous phase reactions with H₂O₂). We don't expect the relative trends (main focus of this study) to be strongly affected by this data selection, unless there is significant, long-term shift in weather regimes (and the associated changes in both cloud and air pollution conditions). For the extremely polluted regions discussed in this study, satellite trends in cloud reflectivity (less than +/-2%/decade (Herman et al., 2013)) are much smaller than those caused by changes in emissions (see section 3). We expect clear sky biases to be more important for unpolluted regions (Geddes et al., 2012). This is certainly a very interesting question, but to fully address it, a lot more data and analyses will be necessary.

- We have added statements in section 2.3 as follows: “We note that the CRF is approximately twice as larger as the effective cloud fraction derived assuming Mixed Lambert-

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Equivalent Reflectivity (MLER) cloud model (Boersma et al., 2011; Bucsela et al., 2013; Stammes et al., 2008). Given the very small CRF thresholds, the remaining cloud related errors were estimated to be less than 20% (Lee et al., 2009; McLinden et al., 2014). However, by selecting mostly clear sky conditions, our sampling of the OMI dataset may introduce a bias relative to all-sky conditions (Geddes et al., 2012; McLinden et al., 2014). Clouds are also associated with certain weather conditions, which in turn may affect the level of pollution. These factors may introduce biases in our derived trends in SO₂ and/or NO₂, but only if there is significant, long-term shift in weather regimes. However, for polluted regions in Fig. 1 satellite derived regional trends in cloud reflectivity (less than +/-2% per decade (Herman et al., 2013)) are much smaller than those caused by changes in emissions (see section 3). “

3) P26569 L26-28 Here and in general elsewhere in the text: it is mentioned that trends in OMI columns match trends reported in emissions: I think should be at least roughly quantified. For example, what would be the expected reduction in SO₂ columns corresponding to the observed emission reduction in eastern US? Please check this for other regions too (if relevant).

- We agree. We have updated Figure 3 adding power plant SO₂ and NO_x emission changes for eastern US (OVR box in Fig.2) and northeast India (Fig.6).

We have added the following text in section 3.1: “Fig. 3 (upper row) compares year-to-year changes in the OMI SO₂ and NO₂ annual columns and bottom-up emissions from power plants over the ORV region (blue box in Fig. 2) with other heavily polluted regions discussed later. Overall, between 2005 and 2015 the SO₂ drop over ORV was close to 80%, while NO₂ dropped by 40%, the largest reductions seen in this study. Previous studies demonstrate a linear ~1:1 relationship between the percent change in NO_x or SO₂ emissions from isolated power plants and the corresponding changes in OMI columns (Fioletov et al., 2011, 2015; de Foy et al., 2015). However, Duncan et al. (2013) show that most power plants, such as in the eastern US, are co-located with mobile NO_x sources, so that this relationship is not always obvious. Indeed, OMI

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observed smaller drop in NO₂ columns (~40%) than would have been expected from ~60% reduction in NO_x emissions from the power plants in the region (Fig. 3). “ We have added following text for India in section 3.4: “During the last decade OMI observed much smaller NO₂ increases (~50%) than one would have expected from the increase in NO_x emissions from the coal-fired power plants (Fig. 3h). One possible explanation for the discrepancy might be relatively high NO₂ background from other emission sources. While coal-fired power plants may be the single largest contributor to SO₂ in this region, transportation is a larger contributor to NO_x, and the slower increase in transportation emissions could have masked the sharp increase in coal-fired power plants NO_x emissions. In India, the prevalence of motorcycles with small, two-stroke engines lead to high transportation emission factors for CO, VOC and PM, but produce only modest amounts of NO_x (Dickerson et al., 2002). Also, with a 3-fold increase in NO_x emissions from the power plants, there could be some non-linear effects in NO_x chemistry, changing the lifetime of NO₂. Heavy loadings of soot may also remove NO₂ (Dickerson et al., 2002). The discrepancies will be addressed in future studies. “

4) P26572 L4 “SO₂ reduction” maybe should be specified reduction of what, e.g. emissions” - Done. We revised the sentence: “Europe experienced a ~80% reduction in SO₂ emissions between 1990 and 2011”.

5) P26572 L6 “OMI detection limit” maybe you could remind the value here: - Done. “. . . OMI detection limit of 0.2 DU”

6) P26573 L8-9 I think there is need for a reference here concerning the emission distribution - Thank you for pointing out missing reference. We have added the following reference: Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B. and Li, M.: HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution, *Atmos. Chem. Phys.*, 15(19),

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11411–11432, doi:10.5194/acp-15-11411-2015, 2015.

Technical corrections: 1) Fig. 3 and elsewhere is there a reason you use DU and molec./cm² for SO₂ and NO₂, respectively? Why not just use molec./cm² for both? - In all operational datasets SO₂ column density is provided in DU, while NO₂ column density is provided in molec./cm². This difference is primarily due to the common usage within the SO₂ and NO₂ communities, the former typically using DU and the latter molec./cm². We provided conversion factor from DU to molec./cm² in section 2.1: 1 DU= 2.69×10¹⁶ molecules cm⁻². - We also marked color bars in fig.2-7 in both DU and molec./cm²

2) Fig. 4-7 The panels are quite small and a colorbar for each panel is not necessary. Could the colorbar be moved to the right side (vertical) of each row? So only 1 colorbar for 3 panels. - We agree. Figures 2, 4-7 have been re-done with only one color bar moved to the right side.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 26555, 2015.

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