Atmos. Chem. Phys. Discuss., 14, C2364–C2371, 2014
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term MAX-DOAS network observations of NO_2 in Russia and Asia (MADRAS) during 2007–2012: instrumentation, elucidation of climatology, and comparisons with OMI satellite observations and global model simulations" *by* Y. Kanaya et al.

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Received and published: 13 May 2014

We thank the reviewer very much for reading our paper carefully and giving us valuable comments. Detailed responses to the comments are given below.

Comment 1: Abstract: Overall, the abstract is the worst part of an otherwise great manuscript. I probably wouldn't have read this paper based on the poor abstract. The

authors are doing themselves a disservice.

1)(line 41): I doubt many will know what an "aerosol shield effect" is. Really you are just saying that aerosols weren't adequately accounted for in the radiative transfer calculation. Please consider rewording this sentence.

2)I would make it clear that this is the first study with MAX-DOAS in Russia and Asia.

3)(line 45): "The prevailing : : : " This sentence doesn't say anything. You know that the sites are located in urban settings.

4)(line 46): "The presence : : : " This sentence is not clear at all. Why "diesel vehicles" and what "other sources"?

Reply 1: The abstract is rewritten. Instead of "aerosol shielding effect" we will use a phrase "incomplete accounting of NO2 near the surface under relatively high aerosol conditions" for the satellite observations. We will also mention that this is the first synthetic data analysis for the MADRAS network observations. The previous sentences "The prevailing seasonal patterns with a wintertime maximum implied the dominance of anthropogenic emissions around our sites. The presence of weekend reductions at Yokosuka and Gwangju suggested the dominance of emissions from diesel vehicles, with significant weekly cycles, whereas the absence of such a reduction at Hefei suggested the importance of other sources." were removed. We now state more generally that "Weekend reduction in the TropoNO2VCD found at Yokosuka and Gwangju was absent at Hefei, implying that the major sources had different weekly variation patterns." One sentence regarding diurnal variations as follows will be added: "While the TropoNO2VCD generally decreased during the midday hours, it exceptionally increased at urban/suburban locations (Yokosuka, Gwangju, and Hefei) during winter."

Comment 2: Section 2.3 Retrieval algorithms & Summary. As you discuss later, some of the discrepancies between the data from MAX-DOAS and OMI are associated with different assumptions used in the two OMI and MAXDOAS retrieval algorithms. Could

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you describe the two OMI retrieval algorithms, say in a new Section 2.4, and highlight the main differences between the two OMI algorithms and the algorithm used for the MAX-DOAS instruments? You could also comment on the limitations of satellite data, such as spatial coverage, which should be accounted for when comparing to surface observations.

Reply 2. As suggested, we will describe the two OMI retrieval algorithms in a new Section 2.4 as follows, highlighting differences in the observational information content regarding aerosols and vertical profiles between MAX-DOAS and satellite observations:

2.4 OMI satellite data products for comparison

In this section OMI satellite data products of TropoNO2VCD, to be compared with our MAX-DOAS products in section 3.1, are summarized. The OMI is a UV/vis nadir viewing spectrometer on the National Aeronautics and Space Administration (NASA) Aura satellite on a sun synchronous orbit launched in 2004. An OMI pixel size is 13 $\times$ 24 km2 or larger. We used two different products, i.e., one derived from the algorithm developed by the NASA and the other from the algorithm (Dutch OMI NO2 (DOMINO)) developed by Koninklijk Nederlands Meteorologisch Instituut. The NASA data set was the ver. 2.1 release of the gridded OMNO2d daily level 3 products (OMNO2d.003), with cloud screening at 30%, at a resolution of 0.25° $\times$ 0.25°, available from the NASA Giovanni website (http://gdata1.sci.gsfc.nasa.gov/daac-bin/G3/gui.cgi?instance_id=omi; Bucsela et al., 2013). The latter data set was the monthly DOMINO ver. 2.0 collection 3, at a resolution of 0.125° $\times$ 0.125°, available from the Tropospheric Emission Monitoring Internet Service (TEMIS) website (http://www.temis.nl/airpollution/no2col/no2regioomimonth_col3.php; Boersma et al., 2011). The data at the nearest grid were used for both products. For the DOMINO algorithm, the results at eight adjacent grids were included (gray lines in Fig. 7) in addition to the nearest grid, to represent the spatial inhomogeneity of NO2 over the range 0.375° $\times$ 0.375°. The two algorithms subtract stratospheric NO2 component

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as simulated by a chemical transport model (DOMINO ver 2.0) or determined directly from satellite data (NASA ver. 2.1) from the total and then determine TropoNO2VCD using tropospheric air mass factors. For both satellite data products, air mass factors were computed as average of clear and cloudy conditions weighted by the cloud radiation fraction, and therein the aerosols are implicitly taken into account similarly to clouds (Boersma et al., 2011; Bucsela et al., 2013). They both adopt the vertical profile shapes of NO2 simulated by global chemical transport models at relatively coarse resolutions (TM4 at $2^\circ \times 3^\circ$ for DOMINO and GMI at $2^\circ \times 2.5^\circ$ for NASA) but with down to monthly time resolution. The uncertainty for individual retrievals of TropoNO2VCD was estimated to be 1.0×1015 molecules $cm-2$ + 25% (Boersma et al., 2011) and on the order of 1015 molecules $cm-2$ (Bucsela et al., 2013) for clear-sky conditions. Observational information content with respect to vertical profiles of NO2 and the amount of aerosols is less than the case of MAX-DOAS observations; advantages of MAX-DOAS are that 1) observations of ΔSCDs of NO2 at multiple axes are available, 2) simultaneous determination of aerosols is enabled using O4 absorbances determined in the same axes, and 3) the determined aerosol quantities are explicitly taken into account in the NO2 retrievals, although data at fixed locations with the instruments are only available.

Comment 3: Section 3.1. It is easy to get a great correlation between OMI and MAX-DOAS since they both will reproduce the seasonal cycle in NO2, which is associated with its chemical lifetime. I recommend deseasonalizing the data for a fairer assessment of how well the MAXDOAS and OMI data agree.

Reply 3: For both MAX-DOAS and OMI, the seasonality in TropoNO2VCD arose from the observed SCDs themselves, without using external information (e.g., a priori modeled seasonality, chemical lifetime etc), although seasonality in the vertical profile SHAPES was only used for OMI. Therefore the shown correlation (e.g., Fig. 8) is not automatically expected and is worth to be discussed. On the other hand, comparison of deseasoned data as suggested by the reviewer is important to test if the satellite

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captures the day-to-day variations. In the revised manuscript, we will add one figure (attached, to be inserted as Fig. 12 in the manuscript) and explanation for it at the last part of Section 3.1 as follows:

Deviations of pixel-level OMI(NASA) TropoNO2VCD from monthly mean values positively correlated with those for coinciding MAX-DOAS observations (Fig. 12, with R2 values of 0.45) with a slope of 0.40, a similar value of those in Fig. 9. This suggested that the satellite observation successfully captured day-to-day variations in addition to the monthly variations, although the sensitivity was consistently small.

Comment 4. Line 465: This paragraph is very unclear. Line 473: Describe how "the effect is already taken into account more adequately." Line 485: Huh? How are the vertical profiles treated differently in the retrieval algorithms?

Reply 4. The Lines 465, 473, and 485 of the submitted manuscript are in the paragraph starting from line 11 of page 2901 of the typeset ACPD manuscript. In the revised manuscript, several unclear sentences in the previous manuscript are revised and more explanation is given. The improved paragraph will be as follows: Another possibility would be that systematic underestimation by satellite observations arises from assumptions in the vertical profiles and aerosol treatment. Figure 10a shows that low OMI(NASA)/MAX-DOAS ratios (using a gridded data set for OMI) are associated with high AODs (as observed by MAX-DOAS); although the median ratio is near unity at low AODs ($\sim$0.1), it becomes lower ($\sim$0.7) with AODs as high as 1. In this study, only data with more than 1 $\times$ 1015 molecules cm$-$2 for both MAX-DOAS and satellite observations are used. This suggests the possibility that the satellite observations underestimate TropoNO2VCD when aerosols are densely present. This is less likely to be explained by overestimation by MAX-DOAS at high AODs, where larger observational information content regarding aerosols (multiple axis measurements of O4) were used in the derivation of Abox and TropoNO2VCD. All of the data (n = 1834 from the six sites) were subdivided into two groups of equal size, based on AOD values (i.e., two groups with high and low AOD values) and a Welch's t-test was applied to test the

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statistical significance of the difference between the two means. The results suggested that the OMI(NASA)/MAX-DOAS ratio was significantly lower for the group with higher AODs at the 95% confidence level. Similar tests for individual sites led to the same conclusion for Fukue, Zvenigorod, and Gwangju.

Figure 10b shows that the OMI(NASA)/MAX-DOAS ratio for TropoNO2VCD had a weak decreasing trend with the retrieved parameter v1, the fraction of NO2 present in the lowest 1 km. The median values decreased from around unity to 0.67 as v1 increased from ∼0.6 to 0.9. Welch's t-tests applied to two groups of data sorted by v1 values suggested that the ratio was significantly lower for the group with higher v1 values when using data from all six sites and when using data from Yokosuka and Hefei individually, at the 95% confidence level. This suggests that the underestimation occurs when NO2 is mostly present near the surface. These analyses, in combination, imply that the lower values from the satellite could be partly caused by the assumptions made regarding the vertical profiles and aerosol treatment in the satellite data analysis. This may be important at clean sites, where the spatial inhomogeneity cannot be responsible for the difference. For both satellite data products, air mass factors were computed as average of clear and cloudy conditions weighted by the cloud radiation fraction, and therein the aerosols are implicitly taken into account similarly to clouds (Boersma et al., 2011; Bucsela et al., 2013). Considering that the variance of the ratio was only partly explained by AOD (Fig. 10a), one could argue that the effect of aerosols was almost successfully removed even in the current satellite data retrieval. However, a weak dependence of the ratio on AOD was still discernible, suggesting that the retrieval could be improved by an explicit treatment of the aerosols. Recently, Shaiganfar et al. (2011) and Ma et al. (2013) suggested that the shielding effect of NO2 by aerosols could be significant for OMI observations, resulting in similarly low values. Lin et al. (2013) suggested that concentration of aerosols at the top of the boundary layer increased retrieved NO2 by 8%.

Comment 5: Figure 3. Why is OMI NO2 much higher over the ocean near Japan than

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over Japan's cities?

Reply 5: The map of the country borderline did not match precisely. An improved figure will be used.

Comment 6: Figure 7. Why are the error bars on the MAX-DOAS data so much larger than those on the OMI data? It seems from your conclusions that the error bars on the OMI data should be much larger.

Reply 6: The major cause of the large bars is that data with relatively large values are included in MAX-DOAS. It should be noted that the bars represent 1-sigma ranges of the MAX-DOAS and satellite data, and do not include systematic uncertainty. Satellite observations sometimes fail to reproduce data with relatively large values, especially when high v1 values are associated. Thus, our conclusion does not contradict this figure.

We thank the reviewer for their comments helping us to improve our manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 2883, 2014.

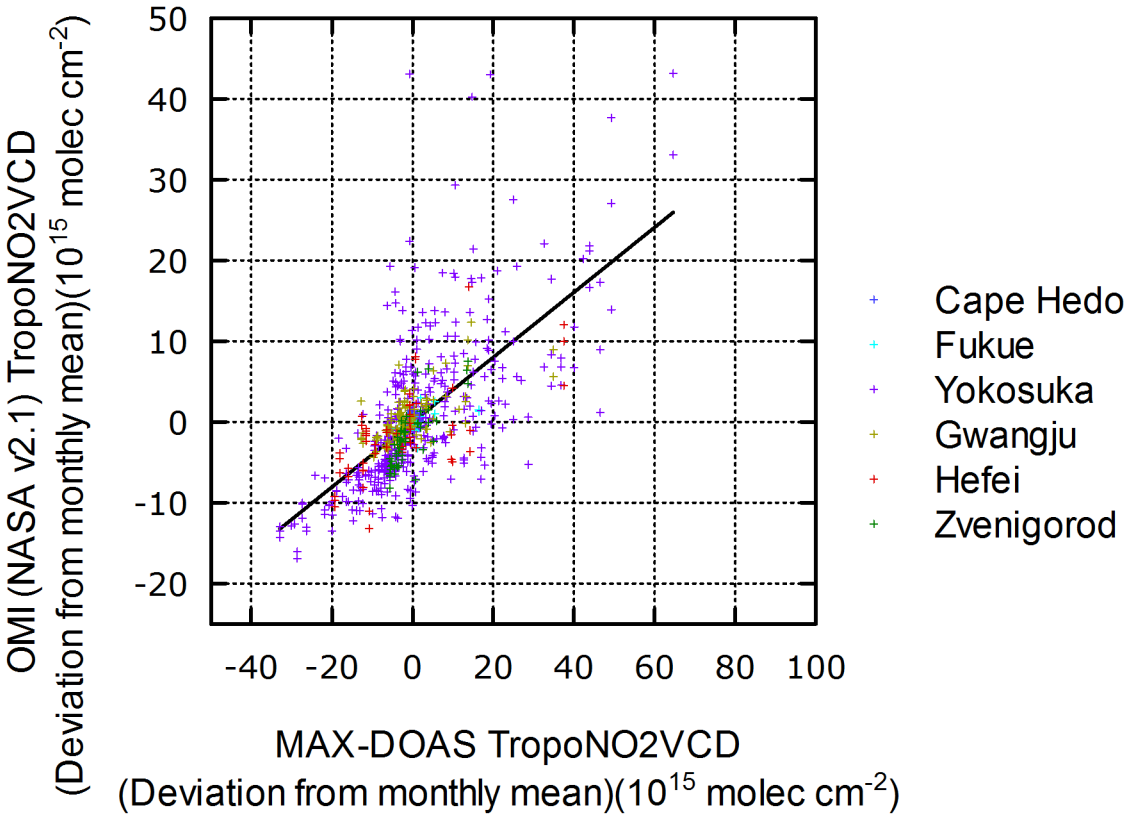

Fig. 1. Deviations of pixel-level OMI(NASA) TropoNO2VCD from monthly mean values positively correlated with those for coinciding MAX-DOAS observations.

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