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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimation of NO_x emissions from Delhi using car MAX-DOAS observations and comparison with OMI satellite data" *by* R. Shaiganfar et al.

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Reply to Reviewer 1

First of all we want to thank this reviewer for the positive assessment of our study and for the many constructive comments and suggestions. We have addressed almost all of these suggestions (for details see below) and have given detailed arguments in cases when we did not follow the reviewers suggestion. Based on the suggestions of this and another anonymous reviewer we have made several important changes to the manuscript, which are briefly summarised in the following: -we derived emission NOx estimates for Delhi also from OMI satellite observations -we considered wind data also

for higher atmospheric layers (up to 1000m) -we explicitly considered the azimuth dependence of the sensitivity of the car MAX-DOAS measurements for the determination of the NOx emissions -we added sensitivity studies for aerosol optical properties and for the effect of elevated aerosol layers

The paper presents Multi-Axis-Differential Optical Absorption Spectroscopy (MAX-DOAS) measurements and derived NOx emission estimates in Delhi and nearby regions. The measurements were made on three days during April 2010 and one day in January 2011. Using simultaneous wind speed and direction data and assuming a constant ratio of NOx and NO2 in the polluted layer, the authors estimated the total NOx emission rate in the considered region. It is found that emission estimates on different days were similar within about 30 percent. The estimation was preceded by an attempt to justify the so called geometrical approximation of the air mass factor. The emission estimates for areas encircled by the measurements were up-scaled to the greater Delhi area and then compared with data of independent emission inventories. Additionally, the paper presents the comparison of MAX-DOAS measurements with tropospheric NO2 vertical column densities (VCDs) retrieved from OMI measurements. The spatial correlation between these data was found to be rather high, but it was found also that OMI VCDs are systematically smaller than those from the MAX-DOAS measurements. I noticed the two following potential major results of this study which could present substantial interest for the atmospheric scientific community and could deserve publication in ACP:

1. Measurement based estimates of NOx emissions in one of the world's largest urban agglomeration;

2. Justification of the geometrical approximation of the air mass factor. Unfortunately, neither of these points is sufficiently elaborated, and further work is needed before the paper can be recommended for publication in ACP.

Major concerns:

1. The method to derive NOx emissions from the MAX DOAS NO2 VCDs involves a pair of factors (c_L and c_tau) defining the assumed NOx to NO2 ratio in the polluted layer and the ratio of the measured NOx and the originally emitted NOx. The authors assume a constant value of 1.32 for c_L with an uncertainty of about 10 percent. However, they note also (p.19188, l. 19-25) that "especially close to strong emission sources, part of the emitted NO might not be quickly converted to NO2 if the NO mixing ratios locally exceed those of O3". Thus the authors seem to recognize that the assumed uncertainty in c_L, and, consequently, the reported uncertainty in the NOx emission estimates may be wrong. It makes me wonder about actual usefulness of one of the major results of this paper. In my understanding, this study provided a rather credible estimate for the lowest limit of the NOx emissions from Delhi on the considered days, while the upper limit is essentially not constrained by the measurements. Therefore I believe that the authors should either clearly specify that they report only the lowest limit of the NOx emissions, or provide additional facts (for example, air pollution monitoring data or urban-scale modeling results, see, e.g. http://urbanemissions.info/model-tools/sim-air/delhi-india.html) which would allow constraining their emission estimates from above.

Author comment: We further investigated the effect of the limited conversion of NO to NO2 close to strong emission sources. In summary, it turned out that such effects are negligible for our study. There are tow main reasons for this: First, according to Gurjar et al. (2004), the relative contribution of power plants to the total NOx emissions of Delhi is far below 10%. Thus the effect of potential limited conversion of NO to NO2 is negligible for our study. We added this information to section 3.1. Second we provide more detailed estimates on the efficiency of dilution during the transport from the power plants to the measurement location (appendix B). These estimates indicate that the potential underestimation of the power plant NOx emissions is negligible for our measurements. We also added this information to section 3.1 (and in more detail to appendix B).

2. The emission estimates derived from MAX DOAS measurements are compared with the estimates of annually averaged emissions from the EDGAR inventory and the study by Gurjar et al. The authors made a warning about inconsistency of the temporal scales, but unfortunately they did provide any hints about possible differences in emission estimates which may be associated with such inconsistency. I recommend the authors to review available information and literature about possible seasonal variability of NOx emissions in India or similar region (e.g., Mexico). In particular, regional emission inventories, as well as seasonal variations specified in different chemistry transport models could be considered and discussed.

Author comment: We agree that it would be very useful to have information about the seasonal variation of the NOx emissions in Delhi. Unfortunately, it seems that such information is currently not available. From measurements of the NOx mixing ratios (e.g. Guttikunda, 2009) a strong seasonal cycle is found with maxima in winter. However, it is not possible to disentangle the seasonality of the corresponding emissions from these time series because: (a) the mixing ratios are also largely determined by variations of the mixing layer height, which has its own seasonal variation. (b) the NOx lifetime also depends on season. Both effects inhibit the use of the surface mixing ratio data to directly quantify the seasonal variation of the NOx emissions. We added this information in section 3.2.

3. The authors estimate possible deviations of true NO2 VCDs from those obtained with the geometrical approximation using a radiative transfer model for various scenarios. The results show that the uncertainties increase with increasing the NO2 layer height, and the authors assume that the NO2 layer height does not exceed 500 m. This is, in my opinion, not a credible assumption: I would expect that NO2 is well mixed within the whole boundary layer, and that the boundary layer height for the considered conditions (at, least, in April) probably exceeded 2000 m. The authors should provide the range of the boundary layer heights (e.g., from the ECMWF analyses used in the study) during the measurements and to show results of sensitivity tests covering this

range. In addition, the deviations should be assessed for the range of possible values of parameters of particle size distribution and the single scattering albedo (SSA). The assumptions regarding SSA and the size distribution should be explained and justified.

Author comment: We agree that the boundary layer height is typically higher than 500m, especially in April. However, recent studies have shown that usually the NO2 layer height especially close to emission sources is systematically lower than the boundary layer and the aerosol layer height. This can be easily understood by the rather short lifetime of NOx (compared to that of aerosols). MAX-DOAS observations in Milan in September 2003 (Wagner et al., 2011) show rather low NO2 layer heights typically below 500m. Interestingly, for HCHO and aerosols systematic higher layer heights were obtained. From this simultaneous retrieval we conclude that the rather low NO2 layer heights are not an artefact, but real. Even for measurements downwind of strong NOx emission sources like at Cabauw, The Netherlands, NO2 layer heights only slightly above 500m were found (Piters et al., 2011). These findings indicate that in general the NO2 profile is systematically lower than the mixing layer height. Thus we assume that at Delhi the NO2 layer is also located systematically below the mixing layer height. We added this information to the revised version of the paper (section 2.5). Inspired by the comment of another reviewer, we applied a first order correction for the azimuth dependence of the measurement (section 2.5). This correction leads to a decrease of the NO2 VCDs by 10% to 16%. For this correction we assumed a NO2 layer height of 500m. This azimuth dependent correction is explained in sections 2.5 and 3.1.

The aerosol optical parameters (Single scattering albedo of 0.95 and asymmetry parameter of 0.68) were chosen according to the study from Dubovik et al. (2002) for urban-industrialised cases. However, as shown in the new sensitivity studies (appendix A) the effect of variations of these parameters is rather small compared to the influence of aerosol optical depth, or NO2 and aerosol layer heights.

4. The authors say (p. 19184, l.15-18) that "the retrieval result represents the differ-

ence of the SCDs : : : and the Fraunhofer regference spectrum" (SCD_Fraunhofer) as defined by Eq. (1). This statement is confusing because just in a few lines below it is said that SCD_Fraunhofer "has to be added" (presumably to DSCD_alfa). Therefore, SCD_Fraunhofer is simply cancelled from Eq. (1), and it is puzzling that it was introduced in the discussion. Actually, the formulations in the preceding study by Wagner et al. (2010) look a bit different. In particular, Wagner et al. suggest deriving VCDs from DSCD_alfa (which is a difference of SCD measured at two angles), rather than directly from SCD_alfa as in this study, and these quantities are obviously not identical. This discrepancy should be explained.

Author comment: The reviewer is right in saying that the 'SCD_Fraunhofer is simply cancelled', but this is exactly what is needed: since from the spectral retrieval only the DSCD is derived, but for the determination of the tropospheric VCD the (absolute) SCD is needed, the SCD_Fraunhofer has to be determined independently and then added to the retrieved DSCD. To make the text more consistent with the procedure described in Wagner et al. (2010) we changed the first sentence of section 2.4 to: 'To determine the SCDïĄą of a measurement, the SCDFraunhofer (together with the change of the stratospheric SCD) has to be added to the DSCDïĄą as discussed earlier by Wagner et al. (2010) and Ibrahim et al. (2010).' As shown in Wagner et al. (2010) not only the (absolute) SCD of the Fraunhofer reference spectrum has to be added to the DSCD(alpha), but also the change in the stratospheric SCD between the time of the Fraunhofer reference spectrum and the actual measurements. Usually, this change is small if the solar zenith angle is below 70°, but for consistency we added the effect of changing SZA.

5. The authors use wind speed data only for 3 lowest levels (0, 30 and 60 m above ground). I guess that the wind speed can be larger at upper altitudes within the boundary layer, and thus the pollution can be transported much more rapidly from the boundary layer over Dehli than it is assumed in the study, leading to an underestimation of emissions. This issue should be clarified, and corresponding uncertainties should be

taken into account.

Author comment: This is a very important point and we thank the reviewer for the hint! Initially we assumed that only winds at the lowest atmospheric levels would be important for the transport of freshly emitted NOx. However, simple calculations taking into account atmospheric stability in Delhi show that the NOx should be quickly transported to higher altitudes than 60m. In the revised version of our paper we considered wind speeds up to 1000m and weighted them by an assumed exponential NOx profile with scale height of 500m. The wind speeds calculated with this approach are between +15% and 40% higher as calculated before. The wind direction also changed slightly (up to 8°). Here it is important to note that the strongest changes in wind speed and direction occur below about 200m altitude. Thus the exact layer height of the assumed NOx profile has only little influence on wind speed and direction. We updated the emission estimates using the new wind data; as expected the emissions increased by about 10 to 30%. However, together with the changes caused by the explicit consideration of the azimuth dependence of the AMF, the increase compared to the original version is only between 10% and 15%. We updated the text in section 3.1 accordingly.

6. Validation of OMI data looks “off- topic” in regards to the main goal of the study (estimation of emissions). There are a number of publications on this subject, but the authors did not even made an attempt to compare their findings with results of other studies. I recommend that either the comparison with OMI data is completely removed from the paper (scientific contribution of this comparison is anyway not clear), or the OMI data are used to get independent NOx emission estimates in the same way as MAX-DOAS data. For me, the secon option is preferrable, and I expect that the second option will not require much of additional work.

Author comment: We agree that the comparison with OMI has a completely different focus compared to the first part of the paper. We followed both recommendations of the reviewer. First we put the results of our validation study into the context of existing validation studies. Second, we added emission estimates derived from OMI observa-

tions for the days for which the emissions were derived from the car MAX-DOAS measurements. Interestingly, existing validation studies show a quite controversial picture: studies based on in situ observations (at the surface or from aircraft) and LIDAR observations indicate that the troposheric NO2 VCD derived from OMI (DOMINO v1.02) is biased high (Boersma et al., 2011, Hains et al., 2010 and reference therein). In contrast, studies based on MAX-DOAS observations (Brinksma et al., 2008; Celarier et al., 2008) indicate that the troposheric NO2 VCD derived from OMI is biased low. The latter results are in agreement with the findings of our validation exercise. We added this information to the revised version (section 4.2).

We added a new section (4.1) about the determination of NOx emission from OMI satellite observations to the revised version of the paper. Although the determination of emissions from satellite observations is in principle straight forward, the associated uncertainties are rather high, mainly because the uncertainties of the (estimated) NOx lifetime are directly proportional to the uncertainties of the retrieved emissions. In total the uncertainty of the determination of NOx emission from OMI satellite observations is of about 70%. The results were added to Fig. 7.

Minor comments:

Abstract, l. 12, “the absolute values show a reasonably good agreement”. This statement contradicts to the next sentence and should be either revised or removed.

Author comment: We removed this sentence and changed the following sentence to: ’Concerning the absolute values, OMI data tends to underestimate the tropospheric NO2 VCDs in regions with high pollution levels. ’

p. 19180: l. 24: Million -> million

Author comment: corrected

p. 19181, l. 7-12: a logical link is missing between the two statements ibid, l. 9: to understand -> for understanding

Author comment: We changed these sentences to: 'The measurements of atmospheric pollutants are important for monitoring the air quality and for understanding the radiative forcing and its impact on climate (Ravishankara et al., 2004; Seinfeld and Pandis, 2006). Currently, still large uncertainties exist with respect to the total emissions of pollutants and their impact on local, regional, and possibly also global scale. The corresponding uncertainties are especially large for many Megacities.'

p. 19182, l.3: confusing usage of the word "superior" Author comment: We replaced 'superior to' by 'advantageous compared to'

p. 19183, l. 2,3: the word "pointing" is used two times in the same sentence. Please consider revising this sentence.

Author comment: The first 'pointing' is replaced by 'mounted'

p. 19185, l. 13: "tropsopheric" -> tropospheric

Author comment: corrected

p. 19190, l. 14: Megacities -> megacities

Author comment: corrected

p. 19191, l. 12, "similar patterns": this statement is at least not evident.

Author comment: We replaced 'similar patterns' by 'the same general distribution of NO2'

p. 19191, l. 26, 27: please provide the average values of OMI and MAX DOAS VCDs together with their uncertainties.

Author comment: We added this information to the text.

p. 19192, l. 9-11: it is not exactly proven in this study that the unknown true VCDs are underestimated by OMI. I would modify the statement by saying "VCDs are LIKELY systematically underestimated by OMI", or it would be even better to talk simply about

differences.

Author comment: We agree with the reviewer that it is initially not clear which data set is closer to reality. The uncertainties of both measurements are different over strongly polluted and less polluted regions. In strongly polluted regions, the uncertainties of the MAX-DOAS observations are rather small because of the low NO2 layer height. In contrast, the uncertainties of the satellite observations are rather high, because the sensitivity decreases strongly towards the surface. In addition, looking in more detail into the assumed NO2 profile for the OMI retrieval above Delhi and surrounding areas, it turns out that a large fraction is located at rather high altitudes (about 50% above 400m), which is partly related to the fact that the profile is an average over a rather large model grid (2 3°). The use of such profiles leads to a systematic overestimation of the true tropospheric AMF and thus to a systematic underestimation of the retrieved tropospheric NO2 VCD (see also Heckel et al., 2011). In contrast, in less polluted regions also the uncertainties of the car MAX-DOAS measurements probably contribute substantially to the differences between both data sets. Because of the higher NO2 layer height the uncertainties of the car MAX-DOAS measurements are larger than over strongly polluted regions. We included this discussion in section 4.2 and in the conclusions of the revised version. We also added more discussion about the assumptions made in the OMI retrieval and the associated uncertainties in section 4.2.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 19179, 2011.