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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 2

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 authors present first measurements in India by Car MAX-DOAS to assess the NOx emission inventory of New Dehli. The paper is generally well written, and meritorious of publication, though some doubts remain about the approach, and effect of aerosols on radiation fields. The paper is generally not placed well in context with recent literature from India, that in part seems to call for a need to reassess the sensitivity studies to bind uncertainty in radiation fields. Other then that, the paper is well written, and well within the scope of ACP.

Author comment: We thank the reviewer for the positive assessment! We added several new references including recent literature from India to the revised version of the manuscript:

Rehman, I. H., Ahmed, T., Praveen, P. S., Kar, A., and Ramanathan, V.: Black carbon emissions from biomass and fossil fuels in rural India, Atmos. Chem. Phys., 11, 7289-7299, doi:10.5194/acp-11-7289-2011, 2011. Satheesh, S. K., Vinoj, V., Moorthy, K. Krishna, Weekly periodicities of aerosol properties observed at an urban location in India, Atmos. Res., 101, 307-313, DOI: 10.1016/j.atmosres.2011.03.003, 2011. Babu, S. S., K. K. Moorthy, R. K. Manchanda, P. R. Sinha, S. K. Satheesh, D. P. Vajja, S. Srinivasan, and V. H. A. Kumar, Free tropospheric black carbon aerosol measurements using high altitude balloon: Do BC layers build "their own homes" up in the atmosphere?, Geophys. Res. Lett., 38, L08803, doi:10.1029/2011GL046654, 2011.

Specific Comments: 1) In Northern India the vertical layering of the atmosphere is anything but simple (Babu et al, 2011; Sateesh et al., 2011). These and other measurements have shown evidence for strong layering of aerosols. By March/April aerosols (and NO2?) reach well above altitudes currently assessed in the paper, and up to 9km.

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How valid is the approximation of geometric AMF under such radiation conditions?

Author comment: Fortunately, high aerosol layers have only a very small effect on the MAX-DOAS AMF. We added detailed radiative transfer studies to the paper (in an appendix) for situations with elevated aerosols (together with additional sensitivity studies as suggested by a second reviewer). We also added the information to section 2.5 that the influence of elevated aerosol layers on the car MAX-DOAS measurements is small.

2) Was the MAX-DOAS used to test AOD assumptions? If so, this is not clear. Is the S/N of the instrument sufficient to use O4 dSCDs to assess radiation fields in a meaningful way?

Author comment: In principle at least limited information about aerosol properties is contained in our MAX-DOAS measurements. However, with current inversion algorithms it is not possible to retrieve the AOD from measurements using only high elevation angles like for the car MAX-DOAS measurements. This potential could in principle be used in future algorithms, but will need considerable development and validation efforts. Thus we did not use the information contained in the O4 observations in our study.

3) The authors measure other gases, i.e., O4, H2O, (Glyoxal?), but do not discuss these measurements further. Could the authors extend Figure 2 to show all the fitted cross-section results, and RMS. Also, some discussion about the variability of these gases in context of error bounds would be useful to illustrate the potential of the technique. What is known, what could be learned?

Author comment: We included the fit results of the additional species to the Fig. 2. For the Delhi car MAX-DOAS measurements, only the absorptions of NO2 and H2O could be analysed with sufficient signal to noise ratio, since for the MAX-DOAS measurements only high elevation angles are used, for which the tropospheric light paths are rather short. Thus for most of the trace gases, the corresponding weak absorptions

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are below the detection limit. We added this information to the text.

4) AOD at 550nm is given as in the range of 0.2-0.5 in one place, and as 0.1 'during afternoons' in another place. Where is which information coming from, and which information applies when, and where? Also, an AOD at 550nm (from MODIS, not available during mornings) is a lower limit for the AOD in the wavelength range used for the spectral retrievals. A higher AOD applies for the discussion of uncertainty, but it remains somewhat unclear what is the basis for estimating the error due to radiative transfer as 20%. For the wavelength dependence of AOD over another Indian City see Sateesh et al., 2011.

Author comment: The aerosol data were taken from the AERONET data synergy tool, http://aeronet.gsfc.nasa.gov/cgi-bin/bamgomas_interactive. We added this information to the text. The range of observed AOD is consistent with those measured by AERONET in January and April 2009 (see http://aeronet.gsfc.nasa.gov/). They are also consistent with those reported in the study of Satheesh et al. (2011) for the city of Bangalore and of Rehman et al. (2011) for the city of Kanpur. We added this information and the reference to the manuscript. Concerning the error estimation, we added further detailed sensitivity studies on the effect of different aerosols to the paper (in the appendix). We also discuss the sensitivity to aerosol optical parameters and elevated layers in more detail in section 2.5.

5) Can the authors add a literature search, or support from their own data, why the assumptions 'aerosols < 2km' and 'NO2 < 1km' are indeed meaningful assumptions for their radiation fields. Based on Figure 3, and taking AOD as 0.5 as the first point shown on this rather extended AOD scale (why up to 3? – this does not appear to connect with the paper), it seems that the error due to azimuth effects alone could be twice as large as the specified error for elevated NO2 and aerosol layers. In particular, with measurements as described here, the azimuth angle continually varies as the result of movements of the sun and the pointing of the sensor (inherent to the 'closed routes' that follow circles). Was any attempt made to account for effects due to the

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changing azimuth? In any event, an expansion of their sensitivity tests towards higher aerosol and NO2 distributions seems desirable.

Author comment: A) Up to now there are not many studies about the NO2 profile height. However, MAX-DOAS observations at the Po valley (Wagner et al., 2011), the Pearl River Delta (Li et al., 2011) and Cabauw (The Netherlands) showed that the NO2 layer is typically shallower than the aerosol layer. This can be understood by the different lifetimes of both species. Generally, the NO2 layer was found to be confined to the lowest 500m or below for measurements close to strong emission sources. Even at some distance of the sources (e.g. at Cabauw) the NO2 layer was systematically lower than the aerosol layer. While for satellite observations this might lead to systematic underestimation of the retrieved tropospheric NO2 VCD, for the analysis of (car) MAX-DOAS observations this is quite advantageous, since the true air mass factors can be better approximated by the geometric approximation if the aerosol layer is above the trace gas layer. We added more information on the relative heights of NO2 and aerosols to the paper.

B) It is true that our sensitivity studies include larger aerosol optical depths than found during the measurements in Delhi. We think this is rather beneficial, since the reader gets a broader idea about the general range of sensitivities. Also other studies at more polluted locations might use the results for the interpretation of their measurements.

C) We want to thank the reviewer for this comment! It is true that the relative azimuth changes systematically while driving closed circles around a city. Thus the deviations of the true AMF from the geometric approximation also change as a function of the position. In principle it is a good suggestion to explicitly consider this dependency for the analysis of the car MAX-DOAS measurements. However, the procedure is complicated and time consuming, and thus we did not apply an explicit azimuth correction for individual measurements in this study. Nevertheless, we followed the suggestion of the reviewer in a slightly simplified way: we calculated true AMFs for the locations of the highest NO2 SCDs. These locations are typically confined to localised parts of the

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driving routes. For the correction of the azimuth dependence we assumed a NO2 layer height of 500m and an aerosol layer height of 2km and an aerosol OD of 0.3. Based on these assumptions we calculated correction factors for the determined NOx emissions, which range from 0.84 to 0.9. (13.4.2010: 0.84, 14.04.2010: 0.9, 15.04.2010: 0.9, 15.01.2011: 0.84). Here it is interesting to note that the correction factors do not change much (less than 5%) if other assumptions were made (NO2 layer height 200m, aerosol layer height 1km, aerosol optical depth: 1). We updated the NOx emissions according to these correction factors in the revised version of the manuscript.

6) Can the authors test the hypothesis whether aerosols and trace gas are collocated? If not, the sensitivity study indicates that an error of 20% could arise alone from the mismatch between aerosol and trace gas vertical profiles that has been tested here over a rather limited parameter space. Higher aerosol and NO2 distributions are likely to increase this error estimate. Some more interpretative discussion of Figure 3, applicable conditions, and resulting uncertainty would be useful.

Author comment: As discussed above (point 5) we expect that the layer height of the NO2 profile is typically located below that of the aerosols.

7) In Figure 5, error bars to indicate absolute uncertainties are missing. The Figure as is creates the wrong impression that the last case study is actually more accurate than previous, while the opposite is the case. Maybe adding two error bars per case study could be a way to illustrate the (rather small) error due to 'data gaps', but it should not be shown without also showing explicit context in terms of the overall uncertainty.

Author comment: We agree and added the total uncertainties to Fig. 5 and 7.

8) Figure 11, Section 4: Uncertain aerosol optical properties, i.e., single scattering albedo, have been shown to strongly affect satellite retrievals at UV wavelengths (up to factor 3!), and add to underestimates of NO2 VCDs from space (Dix et al., 2009). The effect is likely not negligible also for NO2, yet discussion about aerosol optical properties is currently missing in this manuscript. Section 4 should include a brief summary of

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the assumptions made by OMI retrievals (NO2, aerosol vertical distributions, collocation, aerosol optical properties) and discussion should connect with sensitivity tests in Figure 3. A very relevant parameter in this context is single scattering albedo (Barnard et al., 2008; Dix et al., 2009), yet discussion is currently missing in justifying the geometric approximation.

Author comment: We included additional sensitivity studies in the revised version of our manuscript (section 2.5 and appendix A). It turned out that variations of the optical properties (single scattering albedo and asymmetry parameter) of aerosols have a rather small effect on the measurements sensitivity of the car MAX-DOAS measurements compared to other properties like layer heights and aerosol optical depth. We also added more discussion on the assumptions made in the OMI retrievals. Especially over strongly polluted areas the assumed NO2 layer height probably systematically overestimates the true NO2 layer height leading to a systematic underestimation of the true NO2 VCD. In contrast, in less polluted areas, the assumed profile might me more appropriate and the retrieved NO2 VCDs should be much closer to the true values. We added this information to the revised version of the manuscript (in section 4.2).

Literature: Babu S. Suresh; Moorthy K. Krishna; Manchanda Ravi K.; et al. Free tropospheric black carbon aerosol measurements using high altitude balloon: Do BC layers build "their own homes" up in the atmosphere? , GEOPHYSICAL RESEARCH LETTERS, 38, L08803, 2011. DOI: 10.1029/2011GL046654 Barnard, R., R. Volkamer, and E.I. Kassianov. Estimation of the mass absorption cross section of the organic carbon componenet of aerosols in the Mexico City Metropolitan Area (MCMA), 2008, Atmospheric Chemistry and Physics, 8(22), 6665-6679. Dix, B.; Barnard, J. C.; Volkamer, R.; 'Implications of the In-Situ Measured Mass Absorption Cross Section of Organic Aerosols in Mexico City on the Atmospheric Energy Balance, Satellite Retrievals, and Photochemistry' in CURRENT PROBLEMS IN ATMOSPHERIC RADIATION (IRS 2008): Proceedings of the International Radiation Symposium (IRC/IAMAS). AIP Conference Proceedings, Volume 1100, pp. 161-164 (2009). doi:10.1063/1.3116938

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Satheesh S. K.; Vinoj V.; Moorthy K. Krishna; Weekly periodicities of aerosol properties observed at an urban location in India, ATMOSPHERIC RESEARCH Volume: 101 Issue: 1-2 Pages: 307-313, 2011. DOI: 10.1016/j.atmosres.2011.03.003

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Interactive Comment