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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Determination of tropospheric vertical columns of NO_2 and aerosol optical properties in a rural setting using MAX-DOAS" *by* J. D. Halla et al.

Anonymous Referee #1

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The manuscript by Halla et al. describes measurements by a multi-axis Differential Optical Absorption Spectrometer operated at a rural location in southwestern Ontario, Canada, during the Border Air Quality and Meteorology Study. The manuscript gives a detailed description of the experimental setup and the various experimental methods. A combination of data sets was used to constrain the retrievals of vertical column densities (VCD) of NO_2, aerosol optical depth (AOD), and two different measures of boundary layer height from observations of O_4 and NO_2 absorptions in scattered sunlight. The observations are compared to simultaneously performed measurements by LP-DOAS, in-situ NO_2 and PM2.5, aircraft NO_2, as well as satellite retrieved tropospheric NO_2 VCD's and AOD. The authors provide a well thought through

validation of their methods using the aircraft, surface, as well as other observations. The comparison with satellite data, although not extensive, shows that tropospheric VCDs from OMI and SCIAMACHYwere 50% higher than the MAX-DOAS VCD's. A number of special cases are discussed, showing for example the transport of pollution plumes aloft over the site.

This is a well written and very detailed manuscript, worthy of publication in ACP after some revisions. Below are some detailed comments on various parts of the manuscript:

Section 1: The literature in the introduction should be updated. For example, there are newer global NO_x emission estimates than from 1992. Discussion of nighttime chemistry should be removed, as it does not seem to be addressed again in the manuscript.

Section 2: I am confused by the mention of a LIDAR system in the manuscript (for example page 13054, line 20 and page 13056, line 16). This instrument is missing in the experimental description. It appears that the data from this instrument could provide important information for a number of discussions in the manuscript, and in particular for the discussion of the NO_2 profiles and the special cases discussed in Section 4.4.

Section 2.2: Please clarify if the zenith spectrum of each elevation scan was used in the analysis or one single noon zenith scan. In the latter case, how was the possible change in stratospheric VCD dealt with?

Section 2.3: Why was a low temperature (223K) O_3 reference used for the tropospheric

data analysis? Why was water, which has absorptions in this range, not included in the fit?

Section 4: The authors discuss several measures of boundary layer heights (BLH). For example, BLH are determined from the aerosol retrievals, the comparison of surface and VCD NO_2, and various meteorological observations. In principle all these determinations should be very similar. As BLH plays an important role throughout the manuscript I would suggest adding such a comparison to strengthen the manuscript.

Page 13052: As the authors use AERONET data for the comparison, why not used the Angstrom coefficients derived by the AERONET station to extrapolate the OMI and MODIS AOD to the wavelength of the MAX-DOAS?

Equation 9: What boundary layer heights were used in Equation 9?

Page 13059 lines 13-18: I find the comparison of VCD_{Geo} and VCD_{RTM} not very instructive, as VCD_{Geo} was calculated only during times when the geometric approximation is valid. One would thus expect a high degree of correlation between VCD_{Geo} and VCD_{RTM}, solely based on this selection process. This should be formulated more clearly here (it is explained somewhat better in the Conclusions).

Page 13060, lines 26: Why was advection not included as a factor influencing NO_2 levels?

Page 13061 line 8: As the nocturnal and morning NO_2 was elevated and winds were weak at night (see for example Fig 12) I am not convinced that local NO_2 emissions

were not an important part of the NO_2 budget at the measurement site. At least in the morning NO_2 was most likely of local origin.

Page 13064: I cannot see a statistically significant difference between the 2 and 4 degree elevation DSCD. Is it possible that the similarity is due to RT effects? At high aerosol loads the absorption length through the lower troposphere can become quite similar for very small elevation angles. A closer investigation of the O_4 DSCDs at the different elevation angles during this time would allow a better distinction between RT effects and the effect of the NO_2 profile.
An interesting aspect, not discussed in section 4.4.1, is that the 2 and 4 degree DSCDs increase before the other DSCDs and the in-situ observations. This points towards a temporal effect, i.e. an inhomogeneous plume moving into the complex viewing geometry of the MAX-DOAS.

Minor Comments:

Page 13038, line 8: Please use the term “azimuth” instead of “horizontal”

Equation 11: Should this be $dDSCD_i$?

Page 13050, line 20: Isn’t the SCD_{90} subtracted from the SCD_x values and not the other way around?

Page 13069 line 15-16: I do not understand what the cited error refers to.

Figure 6. Please add the extinction errors to panel B. List the slope with error derived by the linear fit.

Figure 8: How was the boundary layer height determined?

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 13035, 2011.