

Author Response to Referees

Responses to Anonymous Referee #1

We thank Referee #1 for his/her comments. Below, we deal with them point by point. The original reviewers comments are included in arial 10 font. Our responses are included below. Text that has been changed or added is in *italics* and the page and line numbers are listed for the new manuscript version that is being submitted. The page and line numbers we refer to in our responses will not be meaningful to online readers although the context of the changes should be obvious.

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 NO₂, aerosol optical depth (AOD), and two different measures of boundary layer height from observations of O₄ and NO₂ absorptions in scattered sunlight. The observations are compared to simultaneously performed measurements by LP-DOAS, in-situ NO₂ and PM_{2.5}, aircraft NO₂, as well as satellite retrieved tropospheric NO₂ 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 SCIAMACHY were 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.

Response: The sentence referencing literature on NO_x emissions sources has been updated to reflect more recent "top-down" approaches at determining global NO_x emissions. We now use the more common unit Tg N yr⁻¹. The % remains unchanged.

Page 3, lines 25-28: *Recent estimates of global emissions of NO_x (NO₂ +NO) to the atmosphere fall in the range of 43 - 46 Tg N yr⁻¹ (Jacob, 1999; Martin et al, 2003, Martin et al, 2006) using both bottom up and top down approaches. Approximately 74% of these emissions are attributable to anthropogenic and biomass burning sources.*

Updated References added:

- Jacob, D.J., Introduction to Atmospheric Chemistry, Princeton University Press, Princeton NJ, 1999.
- Martin, R. V., D. J. Jacob, K. Chance, T. P. Kurosu, P. I. Palmer, and M. J. Evans, Global inventory of nitrogen oxide emissions constrained by space-based observations of NO₂ columns, J. Geophys. Res., 108(D17), 4537, doi:10.1029/2003JD003453, 2003.
- Martin, R. V., C. E. Sioris, K. Chance, T. B. Ryerson, T. H. Bertram, P. J. Wooldridge, R. C. Cohen, J. A. Neuman, A. Swanson, and F. M. Flocke, Evaluation of space-based constraints on global nitrogen oxide emissions with regional aircraft measurements over and downwind of eastern North America, J. Geophys. Res., 111, D15308, doi:10.1029/2005JD006680, 2006.

Section 1: Discussion of nighttime chemistry should be removed, as it does not seem to be addressed again in the manuscript.

Response: Concerning discussion of nighttime chemistry, we argue that a general discussion of major sources and sinks of the main pollutant here (NO₂), and its importance in atmospheric chemistry is warranted. We have modified one sentence to reflect this:

Page 3 lines 35-37: *The main losses of NO₂ in the atmosphere are through the photochemical formation of HNO₃ during the daytime followed by dry and wet deposition, and through the formation of the nitrate radical, NO₃, and dinitrogen pentoxide, N₂O₅, at night.*

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 NO₂ profiles and the special cases discussed in Section 4.4.

Response: A brief description of the LIDAR instrument has been added in Section 2.8 of the paper with reference. In principle, LIDAR could provide important information for a number of discussions in the manuscript, however LIDAR data was only available for limited time periods and was not available for all comparable periods. As such, it was only used for helping to determine the boundary layer for the composite profiles as described in Section 4.2.

Page 9, lines 248-254: *The scanning lidar facility (RASCAL—Rapid Acquisition Scanning Aerosol Lidar), capable of fast azimuth and elevation scanning of the lower troposphere, was present at the Ridgetown site. A full description of its operation is given elsewhere (Strawbridge and Snyder, 2004). It was only used to aid in the determination of boundary layer heights for the composite profiles described in Sec4.2.*

Updated References added:

- Strawbridge, K.B. and Snyder, B.J., Planetary boundary layer height determination during Pacific 2001 using the advantage of a scanning lidar instrument, Atmospheric Environment 38, 5861–5871, 2004

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?

Response: When applying the DOAS fit for NO₂, a single FRS from a clean day was used for the entire field study, in order to obtain NO₂ DSCDs. For O₄, a daily FRS was used to obtain O₄ DSCDs. The stratospheric VCD was dealt with as follows:

For each elevation angle ($\alpha = x = 30, 10, 6, 4, 2$)

$DSCD_x = SCD_x - FRS$ or

$DSCD_{90} = SCD_{90} - FRS$ and thus

$DSCD_x - DSCD_{90} = SCD_x - SCD_{90}$ for a given series with the stratospheric component minimized.

Changes:

page 6, lines 153-154: *All NO₂ fits were performed using a single FRS selected from a clean day during the field study.*

page 6, lines 158-159: *All O₄ fits were performed using daily FRS spectra selected from time periods closest to solar noon.*

page 12, line 352-354: *In order to minimize the effect of stratospheric NO₂, NO₂ DSCD_{meas} ratios were prepared by taking individually measured SCD_x values ($\alpha < 30$) and subtracting from them the SCD₉₀ for a given series, then dividing each DSCD by the DSCD₁₀.*

Section 2.3: Why was a low temperature (223K) O₃ reference used for the tropospheric data analysis? Why was water, which has absorptions in this range, not included in the fit?

Response: A low temperature (223K) O₃ reference was used for the data analysis since most O₃ is found in the stratosphere. Water was not fit in this wavelength range of 410-435 nm because it does not have significant absorptions in this wavelength range.

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 NO₂, 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.

Response: The authors appreciate this suggestion, however we do not think the paper will be improved through a comparison for the following reasons: Several of the parameters, H_{gas} , H_{aer} and BL_{eff} , are not true boundary layer heights and are different from each other. H_{gas} and H_{aer} do not precisely measure the same parameter, as the first is the gas layer height (not necessarily equal to boundary layer height) and the second is the aerosol layer height (also not equal to the boundary layer height). These parameters would only be equal to the boundary layer height under ideal conditions, in which all NO_2 or all aerosols are confined to an ideal box layer. In reality, aerosols and gases in the atmosphere have more complex vertical distributions that are different from one another, and thus do not necessarily agree with one another. The method outlined in the paper has been optimized for the determination of VCDs and AOD, but not necessarily quantitative boundary layer heights. In addition, the true measures of boundary layer height that could be determined at the site using tethered sonde and/or LIDAR observations were very sporadic making a comparison difficult.

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

Response: This is a valid objection and a good suggestion by the referee. However, a quantitative comparison between our AOD determined by MAX-DOAS-RTM, and OMI, MODIS, and AERONET must be treated with caution. While it may be possible to convert the OMI and MODIS AOD to the wavelength of the MAX-DOAS determined by an appropriate Angstrom coefficient, it would be difficult to select this coefficient since both AERONET stations are distant from Ridgetown (~300 km). A rigorous quantitative comparison of the AODs is beyond the scope of this paper for this reason.

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

Response: In equation 9 the aerosol extinction coefficient is calculated from AOD divided by H_{aer} . H_{aer} is defined here as the aerosol layer height. If 100% of the aerosols present are confined to the boundary layer, then $H_{\text{aer}} = \text{BLH}$. The H_{aer} for a given measurement series is determined via Equation 10.

Changes:

page 11, lines 326-327: *where τ_{RTM} is the aerosol optical depth, and H_{aer} the aerosol layer height, determined by the process described by Eq. (10) below.*

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).

Response: The reviewers comments are completely correct, and this will be explained better before the conclusions. We now do this with the following description at the location indicated:

Changes:

page 18 line 561-565: *The selection criteria is quite limiting and is fulfilled for 10 data pairs only, as seen in the table. The criteria has been used in past studies to ensure that the VCDs determined by the geometric approximation are not heavily influenced by aerosols, thus making them appropriate for comparison to satellite measures (Brinkma, 2008; Celarier, 2008). Although our method with full radiative transfer is favored, the VCD_{GEO} comparison allows benchmarking to previous literature.*

Page 13060, lines 26: Why was advection not included as a factor influencing NO₂ levels?

Response: Advection has been added as a 4th factor:

Changes:page 19, lines 603-607: *Four dominant factors ...and (iv) changes in advection patterns."*

Page 13061 line 8: As the nocturnal and morning NO₂ was elevated and winds were weak at night (see for example Fig 12) I am not convinced that local NO₂ emissions were not an important part of the NO₂ budget at the measurement site. At least in the morning NO₂ was most likely of local origin.

Response: The reviewer is correct. Our intent was to imply that there is the absence of a significant transportation source rush hour peak at this site, due to its rural location. We have revised the sentence by removing this statement at the beginning of the sentence:

Page 19, lines 609-611:

As this rural site is not impacted greatly by direct sources of NO_x, the decrease in NO₂ from early morning to afternoon is likely indicative of a combination of increased dilution in a growing boundary layer, as well as increased photoysis of NO₂.

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 O4 DSCDs at the different elevation angles during this time would allow a better distinction between RT effects and the effect of the NO₂ profile.

Response: Upon reviewing, the referee is correct that the difference is not statistically significant. The statement has been modified using some of the referee's language to reflect this. See revised statement after next point.

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.

Response: This is a nice point made by the referee. We have added the referee's suggested statement, almost verbatim in the revised text below. This addresses the current point and the previous one made by the referee, and strengthens the argument made for an elevated plume:

Page 22, lines 691-707:

The results from the MAX-DOAS measurements are particularly informative at this time. In particular, while the DSCDs of NO₂ increased at all elevation angles, the DSCD with $a = 4$, DSCD₄, was marginally higher than the DSCD₂ (though the difference is not statistically significant). This is an observation that rarely occurs, even under high aerosol conditions when the absorption length through the lower atmosphere becomes similar at all elevation angles, making all DSCDs similar. This result can be contrasted to the result early in the morning when the nocturnal boundary layer was still intact. During that early morning period, we observed the typical situation in which DSCD₂ > DSCD₄ > DSCD₆ > DSCD₁₀ > DSCD₃₀, commonly seen when a polluted layer exists at the surface, where larger DSCDs are observed at lower elevation angles due to the larger effective path length of scattered light through the polluted layer (Hönninger et al., 2004). In addition, it can be observed during the time just preceding the pollution plume peak that the DSCD₂ and DSCD₄ increase before the other DSCDs and well before the ground level in-situ observations of NO₂ and SO₂ show any detectable increase. This points towards a temporal effect whereby an inhomogeneous plume moves into the complex viewing geometry of the MAX-DOAS. Both effects strongly suggest that the polluted layer was elevated from the surface, or had higher concentrations above the surface than at ground level (Hönninger et al., 2004). Other evidence that the pollution...

Minor Comments:

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

Response: The word horizontal has been replaced with azimuth.

Page 3, line 55: ...and the azimuth telescope pointing direction, β .

Equation 11: Should this be dDSCDi?

Response: Page 12, line 345, Eq. (11): dDSCD replaced with $dDSCD_i$.

Page 13050, line 20: Isn't the SCD90 subtracted from the SCDx values and not the other way around?

Response: Yes, the words "...them from..." were changed to read "...from them.."

Page 12, line 353-354: ... NO_2 $DSCD_{meas}$ ratios were prepared by taking individually measured SCD_x values ($\alpha \leq 30$) and subtracting FROM THEM the SCD_{90} for a given series, then dividing each $DSCD$ by the $DSCD_{10}$.

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

Response: Error = $VCD_{satellite} - VCD_{RTM}$. Clarification made below for average error

Page 25, lines 819-823:

Intercomparison of satellite instrument derived VCDs with a limited number of comparison points from OMI (N=8) and SCIAMACHY (N=1) indicate that the satellite derived measures were ~50% higher than VCD_{RTM} with a mean error of 0.9×10^{15} molec cm^{-2} for OMI, and with an error of 0.5×10^{15} molecules cm^{-2} for SCIAMACHY.

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

Response: Extinction errors were added to panel B and the slope with error derived by the linear fit was listed in the figure captions. Please note that the slope calculation has also been redone.

Page 42: Slope of the trend line (y-intercept zero forced) = $16 \pm 1 m^2 g^{-1}$, $R^2 = 0.75$.

Figure 8: How was the boundary layer height determined?

Response: The determination of boundary layer height was described in the paper already on page 13054 line 19-21 (ACPD version):

"Boundary layer heights for this time period were determined through a combination of LIDAR, vertical temperature profiles measured by tether sonde at the site, and by aircraft in the vicinity of the site."

and on page line 13055 line 11-13 (ACPD version):

"The boundary layer at this time was 797 ± 45 m a.g.l., determined by potential temperature profiles measured by the aircraft".

Changes:

The following statement was added to the Figure 8 caption in the new manuscript version.

"See text for description of boundary layer height determinations."

Responses to Anonymous Referee #2

We thank Referee #2 for his/her comments. Below, we deal with them point by point.

In this manuscript, results from three weeks of measurements with a MAX-DOAS instrument during the BAQS-Met field study are reported. The instruments and methods used are described, comparisons with other data sets are presented and several case studies are discussed. The paper is overall well written but in parts is too detailed. It reports on interesting measurements and comparisons and contributes to the overall description of the situations during the BAQS-Met field study. I therefore think it could be

published in the BAQS special issue but only after major revisions as discussed below. I have several concerns about this paper:

1) A large part of the manuscript describes the DOAS retrievals and their inversion to vertical columns and aerosol optical depth. However, it is not clear which part of this is really a new development and which is just application of previously developed concepts. My impression is, that the new aspect in this study is the application of the method to three weeks of data and that no really new concepts are presented. I therefore think that this part should be shortened considerably. At the same time, it has also to become clearer as some important aspects remain confusing to the reader, for example if a fixed FRS background was used as stated in the text or if the closest zenith-sky measurement was taken as most of the discussion implies. I'm also surprised that the quantities H_{NO_2} and $H_{aerosol}$ which are introduced in the text are never used later although this would be quite interesting (see detailed comments below).

Response 1a: It is our belief that there is no publication describing a full inversion procedure, such as ours, in order to retrieve NO_2 VCDs using MAX-DOAS DSCDs and radiative transfer modelling with this parameterization approach (aside from Wagner et. al., 2011, that is currently in review). Various groups have presented aerosol retrievals, but have not extended this approach to trace gas retrievals. Also, as referee #2 has pointed out, we have applied our method to real data from a 3-week field study (as opposed to synthetic or modelled data).

Changes 1a: some changes emphasizing the uniqueness of the paper and methodology will be added to the new version of the paper in the abstract, introduction and conclusions.

Response 1b: When applying the DOAS fit for NO_2 , a single FRS from a clean day was used for the entire field study, in order to obtain NO_2 DSCDs. For O_4 , a daily FRS was used to obtain O_4 DSCDs. The stratospheric VCD was dealt with as follows:

For each elevation angle ($\alpha = x = 30, 10, 6, 4, 2$)

$DSCD_x = SCD_x - FRS$ or

$DSCD_{90} = SCD_{90} - FRS$ and thus

$DSCD_x - DSCD_{90} = SCD_x - SCD_{90}$ for a given series with the stratospheric component minimized.

Changes 1b:

page 6, lines 153-154: *All NO_2 fits were performed using a single FRS selected from a clean day during the field study.*

page 6, lines line 158-159: *All O_4 fits were performed using daily FRS spectra selected from time periods closest to solar noon.*

page 12, line 352-354: *In order to minimize the effect of stratospheric NO_2 , NO_2 $DSCD_{meas}$ ratios were prepared by taking individually measured SCD_x values ($\alpha < 30$) and subtracting from them the SCD_{90} for a given series, then dividing each DSCD by the $DSCD_{10}$.*

Response 1c: A discussion of H_{NO_2} and $H_{aerosol}$ did not take place here (although interesting it was not the focus of our interpretation). The authors appreciate this suggestion, however we do not think the paper will be improved through a comparison for the following reasons: 1. Several of the parameters, H_{gas} , H_{aer} and BL_{eff} , are not true boundary layer heights and are different from each other. H_{gas} and H_{aer} do not precisely measure the same parameter, as the first is the gas layer height (not necessarily equal to boundary layer height) and the second is the aerosol layer height (also not equal to the boundary layer height). These parameters would only be equal to the boundary layer height under ideal conditions, in which all NO_2 or all aerosols are confined to an ideal box layer. In reality, aerosols and gases in the atmosphere have more complex vertical distributions that are different from one another, and thus do not necessarily agree with one another. The method outlined in the paper has been optimized for the determination of VCDs and AOD, but not necessarily quantitative boundary layer heights. In addition, the true measures of boundary layer height that could be determined at the site using tethered sonde and/or LIDAR observations were very sporadic making a comparison difficult.

2) The comparison of the retrieved NO₂ VC with satellite data and profiles derived from air-borne observations is discussed in some detail. While the approach taken is sound, the results are very much limited by the small number of coincidences. For SCIAMACHY, only one comparison was possible and even for OMI, only 8 points remained! I don't think that this is contributing significantly to the validation of these data sets, and discussion of the differences to results from previous comparisons is of very limited interest.

Response: Most literature often makes use of the geometrical approximation to convert DSCD-30 to a VCD that is compared to satellite results – although our results are limited due to a small number of coincidences with the satellite measures, we believe this comparison is indeed significant because it is complimentary to what has been previously reported (rural vs polluted), and furthermore it strengthens our argument that radiative transfer modelling should be used to get accurate VCDs as opposed to using a geometrical approximation. While 8 points is not a large number, the statistics of those 8 points are presented, such that the statistical significance of the comparison can be made by the reader themselves.

3) The air-borne profiles used for validation have problems as well – apart from the fact, that they were taken at different distances from the MAX-DOAS instrument for different altitudes and do not cover the interesting altitude region, they also are identical within their scatter for the two profiles shown in Fig. 8. Still, the authors construct two different profiles from these data which I think is not supported by the measurements.

Response: The authors acknowledge that there are sources of uncertainty in the air-borne profiles, i.e. there are limitations in deriving a vertical profile from aircraft legs that are not coincident with the site. These uncertainties have been stated clearly in the paper. However, the profiles shown in Fig. 8 are not identical. The main difference in the AM vs. PM profile is the distribution of NO₂ within the boundary layer. In the morning the NO₂ is assumed to be confined to a shallow boundary layer, while in the afternoon, the same assumption was found to be invalid, and the NO₂ is assumed to decay exponentially from ground level. The concentration at ground level, which can be a major contributor to the column determination IS different for every profile in the AM and PM profiles. Our approach in the construction of the profiles was not to assume they were the same, but to presume they were different, since theoretically, they should be and are in fact different (just that our statistics can't show it). In the end, the total composite VCDs we derive from this analysis (Table 1) are not statistically different from one another (4.06 ± 1.11 ; 3.85 ± 1.19 , 3.06 ± 1.22 , 3.18 ± 1.22), but likely only because we did not have sufficient data to truly show the differences in the profiles. However the fact that they are not statistically different, in our view, is not sufficient evidence to make them the same, since there is sufficient theoretical framework to argue that they are, and must be different. We would prefer to leave the 4 profiles numbers different, as we believe they should be. In the end, it is more accurate this way.

4) In the last part of the manuscript, several case studies are discussed, highlighting the usefulness of MAX-DOAS observations as compared to in-situ or active DOAS measurements. While the study on June 30 is quite straightforward, I do have problems with the vertical NO₂ columns presented for July 2nd and July 9. In both cases, large NO₂ columns are derived in times with little separation of the viewing directions. In particular on July 2nd, a rather small change in DSCD₃₀ (factor of 2) leads to a large change in VCD (factor of 5). In this situation, the DAMF_{xx} appears to be only of the order of 0.2 for all directions, indicating that the sensitivity of the measurements to the NO₂ is very small. How realistic are then the results? Can the enhancement in NO₂ column also be seen in the zenith-sky observations? Could this possibly also be related to the aerosol layer mentioned in the text? I'm also worried by the fact that the AOD is actually reduced during these episodes – if you could see the haze from the fires, AOD should certainly be significantly enhanced!

Response: The case studies have been further examined. For July 2nd, it can be seen that the DSCD₃₀ increased in the late afternoon, and the fitted VCD_{RTM} did as well. It can also be seen that the DSCD₂ is only slightly higher than DSCD₃₀, while the DSCD₉₀ (zenith sky observations) also increases at this time.

This suggests that NO₂ is very high up, in the upper free troposphere likely rather than in the boundary layer. The H_{gas} values for this time period (not shown) are also very high (3-5 km), providing further evidence that the enhanced NO₂ was vertically very high. The authors acknowledge that the interpretation of these results is somewhat limited due to the RTM modelling conditions used (ie- the methodology used here cannot correctly determine a 2 layer system, which is likely what exists here. Furthermore, when looking at the AOD during the periods where the NO₂ VCD_{RTM} rises the AOD actually increases slightly as well (not decreases), albeit ever the increase is small. This modest increase is likely due to the distance that the fires were from the site (>1000km). We will highlight this point as well as other evidence for the distant plumes impacting the area on this day, in the final version of the manuscript.

Details comments:

P 13037, L4: have been estimated

Response: line has been rewritten

Page 3, lines 25-27: *Recent estimates of global emissions of NO_x (NO₂ + NO) to the atmosphere fall in the range of 43 - 46 Tg N yr⁻¹ (Jacob, 1999; Martin et al., 2003, 2006) using both bottom up and top down approaches.*

P 13037, L25: not sure the size of the molecules is the right quantity here – H₂ is small but cannot be measured by DOAS while O₃ can.

Response: reference to small molecules has been removed.

Page 3, lines 46-48: *Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) can also be used for the measurement of certain molecules such as NO₂ and O₄ through the application of the DOAS....*

P 13038, first para: This paragraph is a mix of different things and should be rewritten

Response: the paragraph was split in two at a logical place.

P13038, I17: unlike for active DOAS

Response: changed as suggested.

Page 4, lines 61-62: *...unlike for active DOAS...*

P13041, I28: Here, it is stated that a single FRS was used for all retrievals. Later, this does not appear to be the case. What has been done in the study? What is shown in the figures?

Response - See response 1b above.

P13043, I15 and P13044, I11: no need to reference DOAS again and again

Response: DOAS references removed from the following lines:

P13043, I15; P13044, I11

P13043, I26. How do the assumptions on the NO₂ profile made for the SCIAMACHY retrieval fit to your airborne profiles from Fig. 8?

Response: The a-priori profile used for the SCIAMACHY retrieval has 80% of NO₂ confined in a boundary layer height of 1 km. For the AM airborne profile, approximately 80±5% of NO₂ is within 1.08 km above ground level, and for the PM profile, approximately 78±3% of NO₂ is within 1.10 km above

ground level. In both the AM and PM profile our boundary layer values differ from 1 km, as described in the paper.

P13047, I22 and 13059, I11: I don't see the relevance of the Friess et al. paper here as it does not deal with NO₂ retrievals or the criterion used for the geometrical approximation

Response: Friess et al. paper reference was replaced with Celarier et al., 2008 reference.

Page 10, lines 288-290: *This criterion ensures that the geometrical approximation is valid, and eliminates measurement points greatly affected by horizontal inhomogeneities, aerosols, or clouds (Brinksma et al., 2008, Celarier, 2008).*

Page 18, lines 561-564: *The selection criteria is quite limiting and is fulfilled for 10 data pairs only, as seen in the table. Similar criteria has been used in past studies to ensure that the VCDs determined by the geometric approximation are not heavily influenced by aerosols, thus making them appropriate for comparison to satellite measures (Brinksma et al., 2008; Celarier et al., 2008).*

P13048, I23: most OR predominantly

Response: changed as suggested.

Page 11, line 311: *Since O₄ is predominantly in the lowest part of the troposphere, this...*

P13049, I7: why top of the troposphere and not top of the atmosphere?

Response: changed as suggested.

Page 11, line 320: *...vertical column of the atmosphere from sea level to infinity (top of the atmosphere)...*

P13049, I20: How was the fact that you derive aerosol properties at 360 nm but retrieve NO₂ around 420 nm be treated in the retrieval?

Response: Since there is no useful O₄ absorption band at 430 nm, we had to use 360 nm for our aerosol retrieval. In general the AOD at 430 nm should be smaller than at 360nm. Thus, there is a systematic error in our procedure.

Unfortunately, the Angström coefficient at the measurement site is not known. Thus we cannot simply derive the AOD at 430nm for the AOD derived at 360nm. Moreover, as seen from the AERONET stations at Egbert and Kellog, the Angström coefficient is highly variable in time and space. Nevertheless, the AERONET AOD at 440nm is typically about 80% to 95% of that at 380 nm between June 20 and July 11, 2007. To estimate the systematic error of our procedure, we applied the NO₂ profile retrieval with aerosol profiles scaled with 0.8. These new NO₂ VCDs had an average value of $0.99 \times \text{VCD}_{\text{RTM}}$, showing the difference in wavelength for the different retrievals had, on average, a minor effect, ie – less than 1%.

P13050, I20: I assume that SCD₉₀ was subtracted from SCD_x, not the other way round.

Response: Correct, the words "...them from..." were changed to read "...from them..."

Page 12, line 353-354: *...NO₂ DSCD_{meas} ratios were prepared by taking individually measured SCD_x values ($\alpha \leq 30$) and subtracting from them the SCD₉₀ for a given series, then dividing each DSCD by the DSCD₁₀.*

Section 4.2: Why did you not compare the air-borne NO₂ profile with H_NO₂ derived from the retrieval?

Response: see response to 1c.

Conclusions: First sentence: As stated above, I don't see that you outline a new method in this paper. As it is not new, it should be described only very briefly.

Response: To our knowledge this particular technique has not yet been applied to trace gas inversions. We do realize that many inversion methods are currently used by different groups, but we feel it is still very useful to have more examples of MAX-DOAS inversion, even if this particular method has already been partly applied (to aerosols only). This will be emphasized in the new version of the paper.

Conclusions: Comparison of VC_RTM and VC_geom – you state that the agreement is relatively good but only for those cases where the quality criterion is fulfilled. What about the other cases – how does VC_geom_30 compare to your VC_RTM then?

Response: Within our data there is no clear correlation between VCD_{RTM} and VCD_{GEO} , at times $VCD_{GEO} > VCD_{RTM}$ and at times $VCD_{GEO} < VCD_{RTM}$. As our focus was on using the full radiative transfer procedure to obtain VCD_{RTM} , we provided VCD_{GEO} values mainly for comparison or benchmarking purposes for those who have used it previously.

Table 3: It would be very interesting to add here the meteorological boundary layer height as well as H_{NO_2} as derived from the retrieval.

Response: see response 1c

Figure 3: The box DAMFS O₄ in the “Field Measurements” doesn't make any sense – this should come from the RTM branch above.

Response: The green box labelled DAMFs O₄ in the “Field Measurements” section refer to the DAMFs calculated using the DSCDs measured in the field divided by the approximated O₄ VCD of 1.28×10^{43} molecules²cm⁻⁵ as described in Section 3.2, so the authors consider these DAMFs within the field measurement section (as opposed to the DAMFs O₄ that are calculated from the RTM McArtim in the Forward Modeling section).

Fig. 8: Please use the same scale for both figures. It will then become obvious that the two aircraft profiles are identical within the scatter of values. I don't see good arguments from the data to derive separate NO₂ profiles from them apart from the active DOAS point on the ground.

Response: The scales will be changed in a revised figure. However, as discussed for a previous point, there are good theoretical arguments to NOT assume that the profiles are identical, hence they were treated separately. They may appear similar to the referee, but undoubtedly they are different. We just do not have the precision to determine the difference. Since the VCD_{COMP} were determined independently, there is nothing wrong with reporting them as such, since their uncertainties are reported as well. If there were a theoretical reason why NO₂ profiles should be “identical” at all times, we would agree with the referee.

Fig. 12: Why are there so few NO₂ VC values? There are more AOD values which I find odd.

Response: The reviewer is correct in noting that there are more NO₂ VCD_{RTM} values than AOD values. Since an AOD value is required to ultimately obtain an NO₂ VCD_{RTM} value, each NO₂ VCD_{RTM} must have an AOD value at the same time. This stipulation does not hold true for the converse case. Our NO₂ inversion procedure has further selection criteria. In addition, sometimes the model does not converge and a reasonable result is not obtained.

OTHER CHANGES

A number of other minor changes have been made in response to a separate review of the meteorology sections in this paper by David Sills. Here the page and line number refer to the current ACPD version. The following changes were made :

change: p13053 line 9 – change to “...the boundary layer height is highly...”

pg 13064 lines 12-16: from “~~The maximum of the pollution plume coincided with the arrival of a lake breeze front from Lake Huron to the north. Evidence for this was an increase in the relative humidity, a slight drop in temperature, in addition to results from an observational analysis of all lake breeze fronts and their temporal movement in the study region provided by Sills et al. (2011).~~” **changed to:**

“The maximum of the pollution plume coincided with the arrival of a meteorological feature that had several characteristics of a lake breeze front arriving from Lake Huron to the north. Evidence for this was an increase in the relative humidity, a slight drop in temperature, a subtle, gradual, but discernible shift in wind direction towards the north. It also coincided with arrival from the north of a thin east-west line of cumulus clouds that moved across Ridgetown between 12:00 and 13:00 EDT. The thin line of clouds provides evidence for enhanced lift along the line of this feature, typical of a lake breeze front (Sills, 2011). While final results from an observational analysis by Sills (2011) did not specifically identify a feature with gradients that were sharp enough to be called a lake breeze front (2011), there is consensus that the Ridgetown site was experiencing a Lake Huron lake breeze by 14:00 and that this meteorological feature with enhanced lift preceded the arrival of the lake breeze.”

change old 13065 line 2 to “...with the arrival of the lake breeze. ~~front~~”

change old 13065 line 4 to “...through a process ~~known as~~ similar to fumigation...”

change pg 13065 l 29 , change to “ ...VCD and simultaneous arrival with the lake breeze ~~front~~.”

change pg 13065 l 1-2 “...that can transport pollutants upward (Sills et al., 2011).”

change p13066 15 – “...can thus result in a higher concentration of pollutants at the ~~front~~ leading edge of a lake breeze, that is still not completely understood.”

change p13066 15 – “.....but also the vertical column of pollutants ~~at the front~~ as well.”

change pg 13066 ln 12 - “...temperatures (<21 C) and ~~“classic”~~ low deformation lake breeze ...”

change pg 13067 ln 12 “...high-deformation lake breezes circulations ~~around the lakes~~”

change pg 13067 Ln 12 – “and enhanced turbulence ~~strong convective initiation.~~”

change pg 13067 Ln 17 – “ Ozone (not shown) also recorded ...

change pg 13067 Ln 18 to “Due to the ~~strong convection and~~ cumulus clouds in the vicinity for much...”

change pg 13067 Ln 23 to : “the ~~highly deformed~~ lake breeze front...”

change pg 13068 Ln 1 to : “Michigan, just northwest ~~west~~ of Detroit...”

Acknowledgements: an acknowledgement to David Sills is added.