

We thank the reviewers for their comments and time invested in this thorough review. We also appreciate them recognizing the strengths of the manuscript, including the use of independent measurement techniques and a thorough literature review. We have addressed in detail the reviewer's comments below.

General Comment 1

The title is not clear to me. What is the purpose of this study? The approach followed and results obtained are not revealing any major findings.

Authors' Response to General Comment 1

The authors recognize that the AMTD paper's title may have been ambiguous. The title of the manuscript has been revised to, "*Comparison of tropospheric NO₂ vertical columns in an urban environment using satellite, multi-axis differential optical absorption spectroscopy, and in situ measurements*", which we believe captures the purpose of the study.

We respectfully disagree with the reviewer's assertion that, "The approach followed and results obtained are not revealing any major findings." While some past studies have compared in situ and satellite derived columns, satellite and MAX-DOAS derived columns, or in-situ and MAX-DOAS derived columns, no previous study has compared all three in an urban setting. In this way the study is novel and it has helped to elucidate the similarity and difference between what is measured using these methods.

We have provided a useful advance to the methodology by proposing a simple method to estimate NO₂ columns based on in-situ measurements. While our approach is not without inherent assumptions and limitations, it has demonstrated that NO₂ is not well-mixed within the first 500 m of the troposphere, which suggests that previous vertical columns derived assuming a well-mixed boundary layer likely overestimated the boundary layer's contribution to the tropospheric NO₂ VCD. This useful advance has been more clearly articulated in the paper's discussion and conclusion.

The methodology used in this paper to derive NO₂ columns using in situ measurements also provides an alternative to using a model-derived profile. The authors have revised the introduction of the paper to note that few models can derive a NO₂ vertical profile with a horizontal resolution less than 5 km on a local scale. Furthermore, the use of a local-scale CTM requires a well-characterized emissions inventory (Bechle et al., 2013; Simpson et al., 2012), which currently does not exist for Toronto.

Finally, we have provided some new insight into how the magnitude of the difference between in situ and remotely-sensed VCDs can be interpreted in terms of the horizontal spatial heterogeneity of a region.

General Comment 2

The conclusions drawn here heavily depend upon the method of deriving NO₂ vertical columns from in situ measurements. The method has been applied without testing the validity of the method. The authors could evaluate their method using NO₂ vertical profile from aircraft measurements or a CTM model simulation.

Authors' Response to General Comment 2

The authors have incorporated the following discussion points into Section 2.2 of the revised manuscript:

“The assumption of a vertically decaying NO₂ profile was founded on the following criteria: NO₂ originates at ground level, and undergoes photochemical conversion and dilution primarily through first order elimination processes. Modeling of such a system would yield an exponentially decaying vertical profile. A similar vertically decaying NO₂ profile shape has been observed by Isaac et al., 1998 during summertime aircraft measurements conducted over Egbert, Ontario between the elevations of 600 to 2,900 m (approximately 80 km N of Toronto). These measurements demonstrated that the NO₂ concentration decreases with height with a profile similar to that of the water vapour concentration, a profile that is predominantly associated with the dilution of ground-level emissions. There are only a few studies that have derived NO₂ VCDs from in-situ data without assuming a well-mixed NO₂ vertical profile in the boundary layer. Previous research conducted by Schaub et al., (2006) demonstrated that in situ measurements from ground level up to 3.6 km generally obeyed a hyperbolic profile shape. This shape was applied to extract a NO₂ VCD assuming the NO₂ mixing ratio was 0.02 ppb at 8 km. Overall, the application of Eq. (2) in this paper aligns with the available research that does not rely on a CTM.”

“The uncertainty of the in situ NO₂ VCD was estimated using the fraction of the NO₂ VCD that exceeded 2 km, which for this study was recognized as an average maximum boundary layer height under turbulent mixing conditions based on previous measurements conducted in Southwestern Ontario (Halla et al., 2011). The rationale for this uncertainty estimate is that the NO₂ concentration may have not decayed at a consistent rate within and above the boundary layer. The fraction of the NO₂ VCD contained above 2 km ranged from 0% to 22%, with a median of 3%.”

Regarding the reviewers comment, “The authors could evaluate their method using NO₂ vertical profile from aircraft measurements or a CTM model simulation.” aircraft measurements could not provide NO₂ vertical profile information in an urban environment below 500 m. While our approach is not without inherent assumption and limitations, we believe it provides the best estimate of a tropospheric NO₂ VCD using the available measurements. The authors have revised and improved the following discussion regarding the use of a CTM in the manuscript's introduction:

“Previous studies have assessed the relationship between ground-based and satellite tropospheric NO₂ VCDs, and established that VCDs derived from localised-point measurements can directly capture near-source emissions of NO₂, while the large spatial footprint ($\geq 312 \text{ km}^2$) of satellite measurements make it challenging to retrieve this information (Boersma et al., 2009; Ordóñez et al. 2006; Petritoli et al. 2004). These studies have relied on extrapolating ground-based NO₂ measurements to derive VCDs, either by assuming a well-mixed planetary boundary layer, or using a time-intensive global chemical transport model (CTM) with coarse resolution ($> 250 \text{ km}^2$) to derive a vertical NO₂ profile.

The use of Toronto's NO_x monitoring stations situated at 0.01 and 0.5 km above ground level to derive tropospheric vertical profiles of NO₂ in a near-road environment offered the opportunity to demonstrate that NO₂ is not necessarily well-mixed within 500 m above ground level. These monitoring stations enabled the retrieval of a NO₂ vertical profile with a horizontal resolution of 2.3 km, which is advantageous in comparison to using a CTM, since few models can derive a NO₂ vertical profile with a horizontal resolution less than 5 km on a local scale. Furthermore, the use of a local-scale CTM requires highly spatially resolved emissions and meteorology information (Bechle et al., 2013; Simpson

et al., 2012), which is typically not available for cities. Studies relying on a local-scale CTM, have still used simplifying assumptions regarding the free tropospheric quantity of NO₂, which has previously been assumed to be constant at 0.02 ppb above 2 km (Bechle et al., 2013). ”

General Comment 3

The authors are using outdated version 1.02 OMI NO₂ retrievals while new retrievals have been made publicly available for more than a year. Some comments on differences in results with the v2.0 data have been included, but such subjective statements are making the manuscript weaker. Although 20% difference between v2.0 and 1.02 have been reported in the mean sense, there must be a large spatial variation in the difference given the nature of improvements made from v1.02 to v2.0 in the DOMINO product. The analyses should be revised using current improved retrievals.

Authors' Response to General Comment 3

As recommended by the reviewer, the authors have updated the OMI retrieval data using DOMINO v 2.0. The authors have applied all criteria in Table 5 of the AMTD paper (Table 4 of the revised manuscript) using OMI DOMINO version 2.0 data, as shown below in Figure 5, and intend to include this figure in the revised manuscript. The authors have also provided the relationship between OMI (DOMINO version 2.0) and MAX-DOAS tropospheric NO₂ VCDs, as shown below in Figure 7. Moving to DOMINO version 2.0 data changed a few of the days available and slightly increased the overall number.

The authors have updated their discussion in Section 3.3 as follows:

“Figure 5 shows the linear regression of the OMI (DOMINO version 2.0) versus in situ tropospheric NO₂ VCD. Linear regression was performed using data meeting OMI overpass criteria in Table 4. The OMI tropospheric NO₂ VCD exhibits a negative bias of 58% when compared to the in situ NO₂ VCD and good correlation, with a Pearson R value of 0.81.

Figure 7 shows the linear regression results of OMI (DOMINO version 2.0) versus MAX-DOAS tropospheric NO₂ VCDs. A good agreement is seen between the measurement techniques, which have a Pearson R = 0.80. The slope presented in Fig. 7 derived using linear regression is 0.93 ± 0.11 , or if weighted orthogonal distance regression is applied, this slope is 0.74 ± 0.16 . These slopes are in agreement when their respective errors are considered, and in both cases the slope is not significantly different from 1. Of the 11 available comparison days, 8 OMI and MAX-DOAS measurements agree when the respective uncertainties of each measurement are considered. These results suggest both measurements represent a similar spatial region.”

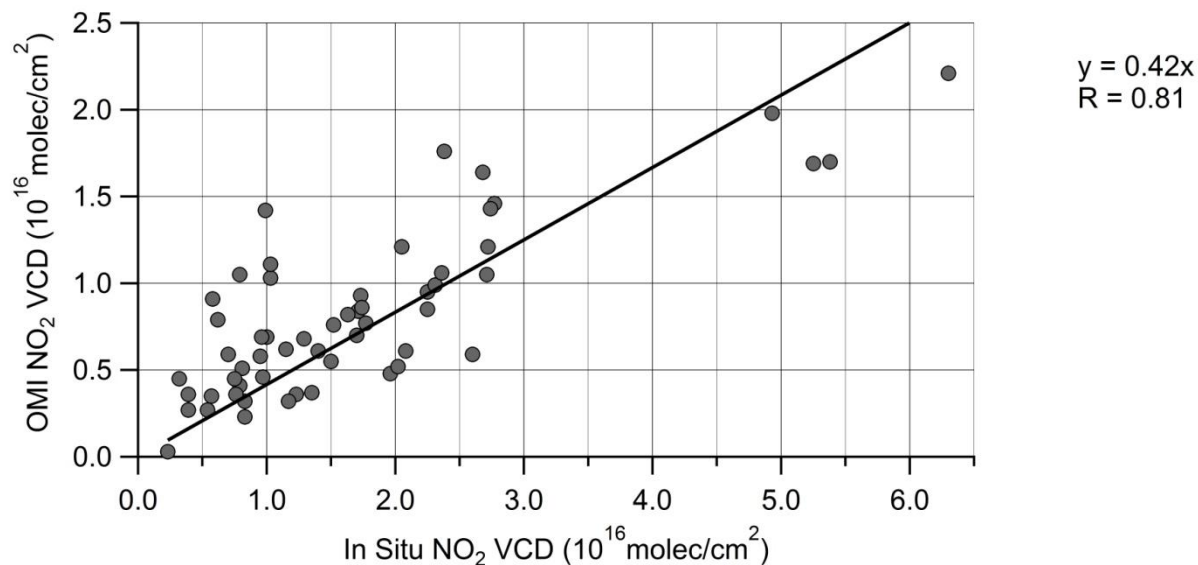


Figure 5. Linear regression of OMI versus in-situ tropospheric NO₂ VCD for data meeting OMI overpass criteria 1 – 4 in Table 4 (n = 56).

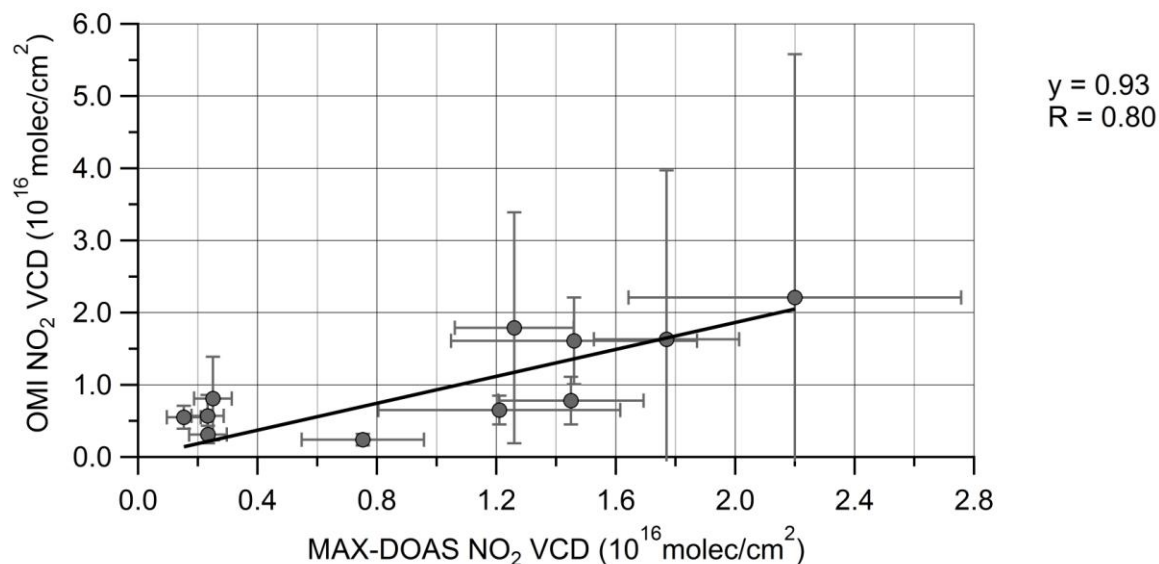


Figure 7. Linear regression of OMI versus MAX-DOAS tropospheric NO₂ VCD (n = 11).

General Comment 4

MAX-DOAS observations are too few (9) to be helpful in drawing any conclusions. Moreover, these observations were taken at different location, elevation, and azimuth angles. I clearly see a danger in interpretation made with poorly characterized data.

Authors' Response to General Comment 4

Authors' Response to Anonymous Referee #2

The authors respectfully disagree that the MAX-DOAS observations are too few (9) to be helpful in drawing any conclusions. As the authors' have discussed in their response to General Comment 3, 8 of the 11 MAX-DOAS measurements agree with the OMI measurements when the errors associated with each measurement type are considered.

The potential error introduced by using MAX-DOAS measurements conducted at different azimuth angles, is described in lines 8 -14 on page 840 of the AMTD paper:

“Shaiganfar et al., 2011 demonstrated that geometric NO₂ VCDs at elevation angles of 22° and 30° differed from those determined using McArtim by $\pm 20\%$ when assuming a 500 m vertical NO₂ box profile ($AOD < 1$). The geometric NO₂ VCD also exhibited a systematic dependency on relative azimuth angle (difference between the solar azimuth angle and MAX-DOAS viewing direction): the geometric VCD underestimated the true VCD at low relative azimuth angles ($\sim 0^\circ$) and overestimated the VCD at higher relative azimuth angles ($\geq 90^\circ$).”

These errors were accounted for in the uncertainty estimate of the geometric NO₂ VCD, as discussed on page 841 lines 4-5 of the AMTD paper.

Regarding the use MAX-DOAS measurements conducted at different locations, as stated in lines 9 -14 on page 830 of the AMTD paper:

“MAX-DOAS draws upon the advantages of both localised-point and satellite-based measurements, offering average pollutant concentrations covering a horizontal scale which has been reported to vary from 3 to 11 km ($\lambda = 357$ nm, aerosol extinction coefficient within 1 km above ground level varied from 1.02 to 0.05 km⁻¹) with a time resolution on the scale of a few minutes (Irie et al., 2011).”

The horizontal resolution of the MAX-DOAS instrument is typically greater than that of OMI. Also, all MAX-DOAS measurement sites were located within 5 km of each other - well contained within the footprint of an OMI pixel, which can reach 13 x 24 km at the finest resolution.

General Comment 5

Presentation of the manuscript should be substantially improved. In few places, there are some lengthy discussions that are not helpful for the intent of this study, but are just distracting. For example, some parts of the first paragraph and 2nd paragraph (Page 827, lines 5-28+) are not adding any value to motivate this study. I do not understand what the authors are trying to say by discussing the results of Petritoli et al and Ordonez et al (in Page 829). Rather than discussing others works, state clearly and concisely how your work is different, superior, and valuable. The paper will benefit by reducing the introduction section and focusing on objectives of the paper. Same comment applies for discussions of MAX-DOAS instrument, retrieval method, and uncertainties. Please, provide the information in consistent manner as in Section 2.4.

Authors' Response to General Comment 5

Lines 1 to 15 on page 827 have been removed. Lines 16 to 28 on page 827 have been maintained since they highlight the utility and need for conducting NO_x measurements in a near-road environment. One of the predominant objectives of this study was to assess the change in the NO₂ column measured near-road using in situ data versus using remotely sensed data with a horizontal resolution > 10 km.

Authors' Response to Anonymous Referee #2

The authors have revised Page 829 in response to Specific Comment #4 from the reviewer. Please see the authors' response to Specific Comment #4 for the revised text.

The authors have removed lines 24 – 28 on page 835 and lines 1 – 6 on page 836 of the discussion paper. The authors have maintained their discussion of the MAX-DOAS retrieval algorithm and software used, since this has yet to be performed consistently amongst all instruments, and therefore, could not simply be referenced.

The authors have maintained their discussion in Section 2.3 of the AMTD paper of the systematic and random errors associated with MAX-DOAS NO₂ VCDs, as although systematic errors associated with the uncertainty, and temperature-dependence of the NO₂ absorbance cross-section have independently been addressed in previous studies, as has the uncertainty of the geometric Δ AMF, they have yet to be consolidated in one paper. These previous studies served as the foundation for the methodology selected in this paper, and as a result, are specifically relevant for inclusion in the paper.

Specific Comments

Specific Comment 1

Abstract: I think, the first sentence is not quite right. The authors did not retrieve trop NO₂ from OMI for the first time. Satellite data did exist for Toronto for a long time.

Authors' Response to Specific Comment 1

The authors can see how the wording “retrieved for the first time in Toronto, Canada using three methods” could be misleading. The intention was to highlight the novelty of comparing three different methods, rather than retrieving data for any one of the methods (e.g. OMI). This sentence has been revised as follows:

“Tropospheric NO₂ vertical column densities were determined for the first time using three methods of differing spatial scales. These methods were compared using data from Toronto, Canada.”

Specific Comment 2

Page 828, lines 4-8: This statement is unclear. NO₂ is retrieved from spectral fit in 405-465 nm window. There is no need to include the wavelength range here as the information is also in the OMI section.

Authors' Response to Specific Comment 2

The authors have re-worded these lines as follows:

“The Ozone Monitoring Instrument (OMI) was launched onboard NASA's EOS-Aura satellite on July 15, 2004, and provides daily tropospheric NO₂ column measurements with a spatial resolution up to 13×24 km² at nadir. OMI has a local ascending equatorial crossing time of 13:45 (Levelt et al., 2006).”

Specific Comment 3

Page 828, line 28: Remove negative sign from longitude coordinate.

Authors' Response to Specific Comment 3

The authors have corrected this typographical error, as shown below:

“(43.663°N, 79.388°W and 43.642°N, 79.387°W)”

Specific Comment 4

Page 829, lines 6-29: What message do the authors want to provide from these paragraphs? Are the authors implying that the methods they intend to use have previously been used?

Authors' Response to Specific Comment 4

The authors intended to comment on the methods that have been previously been used, and have highlighted their limitations. These lines have been revised as follows:

“Previous studies have assessed the relationship between ground-based and satellite tropospheric NO₂ VCDs, and established that VCDs derived from localised-point measurements can directly capture near-source emissions of NO₂, while the large spatial footprint ($\geq 312 \text{ km}^2$) of satellite measurements make it challenging to retrieve this information (Boersma et al., 2009; Ordóñez et al. 2006; Petritoli et al. 2004). These studies have relied on extrapolating ground-based NO₂ measurements to derive VCDs, either by assuming a well-mixed planetary boundary layer, or using a time-intensive global chemical transport model (CTM) with coarse resolution ($> 250 \text{ km}^2$) to derive a vertical NO₂ profile.

The use of Toronto's NO_x monitoring stations situated at 0.01 and 0.5 km above ground level to derive tropospheric vertical profiles of NO₂ in a near-road environment offered the opportunity to demonstrate that NO₂ is not well-mixed within 500 m above ground level. These monitoring stations enabled the retrieval of a NO₂ vertical profile with a horizontal resolution of 2.3 km, which is advantageous in comparison to using a CTM, since few models can derive a NO₂ vertical profile with a horizontal resolution less than 5 km on a local scale. Furthermore, the use of a local-scale CTM requires a well-characterized emissions inventory (Bechle et al., 2013; Simpson et al., 2012), which currently does not exist for Toronto. Studies relying on a local-scale CTM, have still used simplifying assumptions regarding the free tropospheric quantity of NO₂, which has previously been assumed to be constant at 0.02 ppb above 2 km (Bechle et al., 2013).”

Specific Comment 5

Page 830-831: I do not understand what the authors are trying to say here. The discussions sound like MAX-DOAS measurements have previously been used for evaluating satellite retrievals, but there are several difficulties associated with the approach. Then, why authors are still considering the same approach?

Authors' Response to Specific Comment 5

In pages 830 – 831, the authors assessed the previous methodology utilized, and used it to guide the methodology chosen in this paper.

Page 831 lines 5 – 24 have been re-written as follows:

Authors' Response to Anonymous Referee #2

“Previous studies have also investigated the use of stricter coincidence criterion when comparing satellite and ground-based remote sensing techniques as a means to overcome the differences in spatial resolution of the measurements, and improve their agreement. These criteria have included averaging MAX-DOAS VCDs over several azimuths (Brinksma et al. 2008; Celarier et al., 2008), only including OMI measurements with a pixel centre within $0.1^\circ \times 0.1^\circ$ of the MAX-DOAS measurement site (Irie et al. 2008), and excluding MAX-DOAS measurements with a relative uncertainty $\geq 10\%$ (Vlemmix et al. 2010). These studies demonstrated that using stricter coincidence criteria improves the agreement between brings MAX-DOAS and OMI measurements.”

Specific Comment 6

Page 831, lines: 26-28: “The goal of this study was to evaluate. . .”. I do not think this study is evaluating the response of NO₂ column to NO₂ emissions.

Authors' Response to Specific Comment 6

The authors have revised this sentence as follows:

“The goal of this study was to evaluate the response of remotely-sensed NO₂ vertical column measurements in regions with spatially heterogeneous emissions of NO₂, which were examined using in situ-derived column data.”

Specific Comment 7

Page 832, lines 8-12: This information is behind in MAX-DOAS information. I suggest to delete the statement.

Authors' Response to Specific Comment 7

The authors have maintained the following statement, since the use of the geometric AMF is thoroughly addressed in pages 839 – 841 of the AMTD paper:

“MAX-DOAS NO₂ differential slant column densities (Δ SCDs) were converted to tropospheric vertical column densities (VCDs) using the geometric Air Mass Factor (AMF) approximation (Hönninger et al., 2004) in conjunction with the single-scattering validation criteria discussed by Halla et al. (2011) and Brinksma et al. (2008).”

Specific Comment 8

Page 832, line 24: Remove “pollution”.

Authors' Response to Specific Comment 8

The authors have removed “pollution”.

Specific Comment 9

Page 833, lines 11-13: “Both MAX-DOAS and in situ measurements . . .”. I wonder how that would be possible. Please, elaborate.

Authors' Response to Specific Comment 9

The authors have revised these lines as follows:

“The in situ measurements of NO₂ provided the opportunity to observe the impact of diurnal traffic patterns and associated meteorology on the spatio-temporal distribution of NO₂ in an urban environment”.

To address a point raised by another reviewer, the authors have added diurnal trends of the NO₂ mixing ratio (at DT and CN) and the in situ NO₂ VCD for the fall and winter months of December to March, and the spring and summer months of May to August, as shown in Figure 4 below. Figure 4 a, b, d, and f show a peak that corresponds with the morning rush hour periods, and seasonal differences are noted in the magnitude of the NO₂ mixing ratio and VCD at both elevations.

The authors have added the following discussion points in Section 3.2 of the revised manuscript to strengthen the methodology applied in Section 2.2:

“Figure 4 (a) provides the diurnal variation of the NO₂ concentration measured at DT and CN during the fall and winter months of December – March (average temperature = $-0.6 \pm 0.2^\circ\text{C}$), while Fig. 4 (b) shows this diurnal variation during the spring and summer months of May – August (average temperature = $21.1 \pm 0.2^\circ\text{C}$). In both figures, the NO₂ concentration at DT exhibits a peak during the morning hours that corresponds to the evolution of the rush-hour traffic period from 7:00 – 9:00 EST, and decreases during the afternoon. The afternoon decrease is attributed to the increase in planetary boundary layer height, and the increased photolysis rate of NO₂ to produce O₃. The average O₃ concentration (not shown) also reached a maximum during the afternoon hours. The average O₃ concentration at DT during 13:00 – 15:00 EST in December – March was 27.9 ± 1.2 ppb, while in May – August it was 41.7 ± 1.8 ppb. These hours generally corresponded to when the NO₂ concentration at DT reached a minimum.

During December – March at 17:00 EST, the NO₂ concentration at DT began to increase in conjunction with the evening rush hour period, and decreasing planetary boundary layer height. During May – August, the NO₂ concentration remained relatively constant from 13:00 – 19:00 EST, as the higher boundary layer height during warmer months likely resulted in turbulent mixing, and in turn, dilution of emissions during the evening rush hour period. The NO₂ concentration at DT was an average of $25 \pm 1\%$ (5.6 ± 1 ppb) lower in the morning, and $52 \pm 2\%$ (4.0 ± 0.2 ppb) lower in the afternoon/evening during the warmer months than during the colder months.

The NO₂ concentration at CN in Fig. 4 (a) and (b) follows a similar profile as the NO₂ concentration at DT. In both figures, the morning rush hour peak is not as pronounced as at DT, likely due to the vertical dilution and photochemical conversion of NO₂ to O₃ with increasing altitude. The NO₂ concentration at CN also reaches a minimum during the afternoon when the O₃ concentration at CN reaches a maximum. The average O₃ concentration at CN during 13:00 – 15:00 EST in December – March was 33.2 ± 1.1 ppb (19% greater than at DT), while in May – August it was 47.8 ± 4.7 ppb (15% greater than at DT). The NO₂ concentration at CN was an average of $27 \pm 8\%$ (2.4 ± 0.2 ppb) lower in the morning, and $52 \pm 2\%$ (4.0 ± 0.1 ppb) lower in the afternoon/evening during the warmer months than during the colder months.

“Figure 4 (c) shows the diurnal variation of the NO₂ characteristic height, and $[\text{NO}_2]_{\text{CN}}/[\text{NO}_2]_{\text{DT}}$ during the months of December – March, while Fig. 4 (d) shows this diurnal variation during May –

August. Both H_{NO_2} and $[\text{NO}_2]_{\text{CN}}/[\text{NO}_2]_{\text{DT}}$ follow a similar profile since $[\text{NO}_2]_{\text{CN}}/[\text{NO}_2]_{\text{DT}}$ was used to derive H_{NO_2} . As shown in Eq. (2), an increase in $[\text{NO}_2]_{\text{CN}}/[\text{NO}_2]_{\text{DT}}$ results in an increased H_{NO_2} .

The NO_2 characteristic height during the months of December – March exhibited a diurnal trend that is similar to the evolution of the atmospheric mixing height, which increases during the morning hours and peaks in the afternoon (Jacob, 1999). The NO_2 characteristic height (and $[\text{NO}_2]_{\text{CN}}/[\text{NO}_2]_{\text{DT}}$) is an average of 0.5 ± 0.3 km (0.39 ± 0.02) during the hours of 7:00 – 10:00 EST, and reaches an average of 0.65 ± 0.01 km (0.47 ± 0.01) during the hours of 11:00 – 17:00.

During the months of May – August, a statistically significant difference for both the hourly averaged H_{NO_2} and $[\text{NO}_2]_{\text{CN}}/[\text{NO}_2]_{\text{DT}}$ was not witnessed between the hours of 8:00 to 19:00 EST. The average characteristic height during these hours was 0.56 ± 0.01 km – 14% lower than the average maximum characteristic height in December – March, while the average $[\text{NO}_2]_{\text{CN}}/[\text{NO}_2]_{\text{DT}}$ during these hours was 0.41 ± 0.01 – 13% lower than the average maximum ratio in December – March. These results demonstrate that the vertical distribution of NO_2 within 0.5 km during the spring – summer does not follow the typical diurnal profile of the convective boundary layer, which is typically higher during warmer months than colder months.

Previous research using radiosonde measurements has demonstrated that the atmospheric mixing height in southern Ontario (43.682°N , 79.612°W) is an average of 1.5 km during the summer. An increased mixing height during the summer is directly related to the increase in solar radiation and surface heat flux (Ning et al., 1986). The NO_2 characteristic heights shown in Fig. 4 (d) are over 60% lower than these radiosonde measurements of atmospheric mixing height, which suggest they cannot be considered representative of the diurnal evolution of the atmospheric boundary layer height. Overall, Fig. 4 (d) shows that assuming a well-mixed NO_2 concentration within the planetary boundary layer will lead to an over-estimate of the NO_2 VCD.

Figure 4 (e) provides the diurnal variation of the in situ NO_2 VCD during the months of December – March, while Fig. 4 (f) shows this diurnal variation during May – August. The diurnal profile shown in both figures closely replicate the diurnal profile of the NO_2 concentration at DT ($R = 0.87$ and $R = 0.93$, respectively). During the afternoon/evening hours, the NO_2 VCD is at least 2 times greater during December – March versus during May – August. This is due to both the increased concentration of NO_2 DT and the increased $[\text{NO}_2]_{\text{CN}}/[\text{NO}_2]_{\text{DT}}$ during the colder months.”

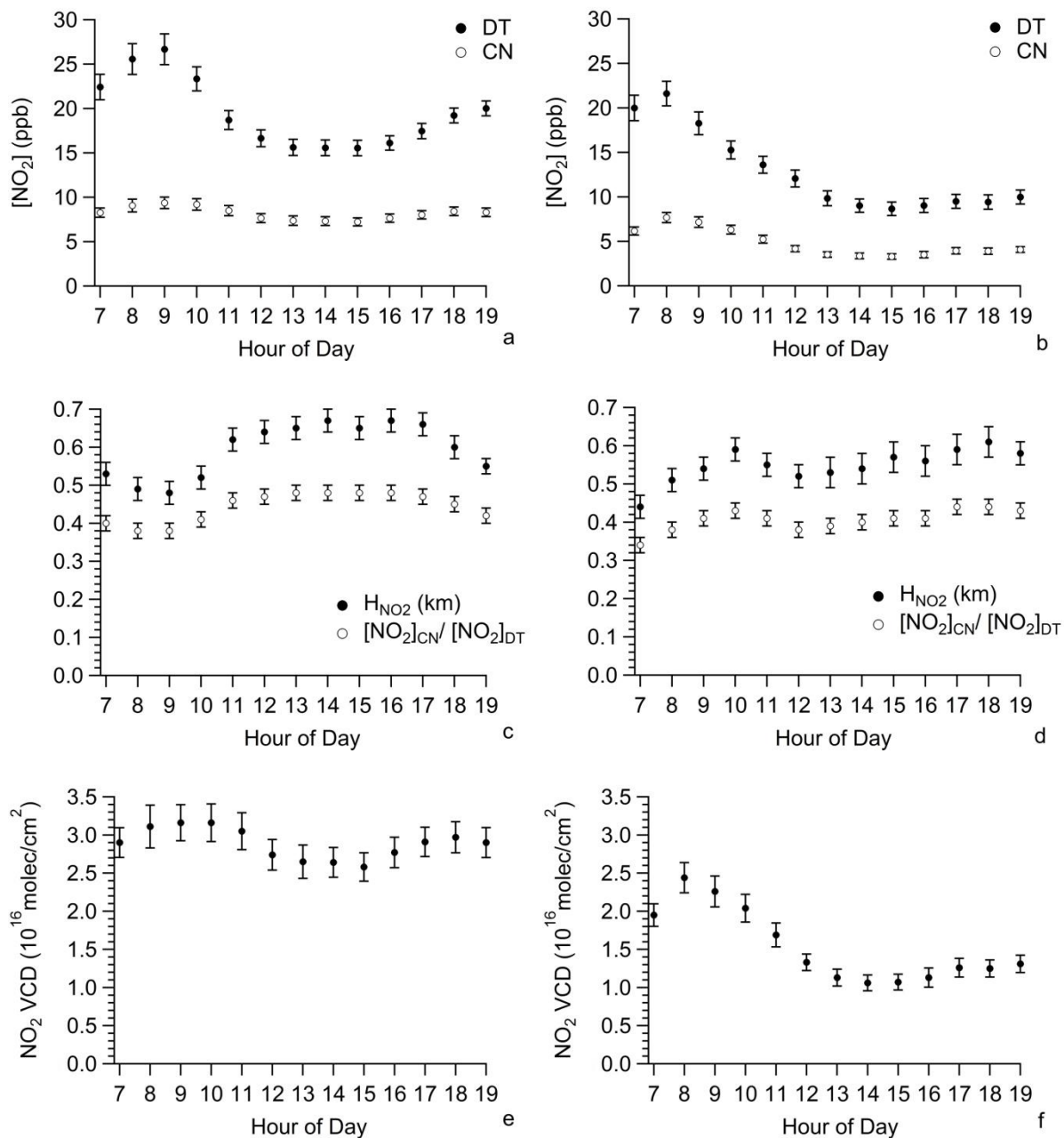


Figure 4 (a) Diurnal profile of [NO₂] at DT and CN during Dec. – Mar. and (b) during May – Aug. (c) Diurnal profile of H_{NO2} and [NO₂]_{CN}/[NO₂]_{DT} during Dec. – Mar. and (d) during May – Aug. (e) Diurnal profile of NO₂ VCD during Dec. – Mar. and (f) during May – Aug.

Specific Comment 10

Page 833, lines 22-23: What is the need of averaging data for 2 hours to compare with OMI and MAX-DOAS observations? Wouldn't it make more sense to use in situ observation that is close to OMI measurement time, which is available in OMI data files?

Authors' Response to Specific Comment 10

Previous studies have averaged in situ measurements over 12:00 – 15:00 LT for comparison with satellite measurements (Bechle et al., 2013), and have yielded conclusive results.

The authors averaged 2 hours of data to increase the efficiency of processing the available data without compromising the results obtained. The NO₂ concentration at CN and DT reached a minimum during these hours, and on average, a statistically significant difference in NO₂ concentration was not observed between measurements conducted at 12:00, 13:00, and 14:00.

Specific Comment 11

Page 834, Eqn 3: This equation is misleading because not all NO_x are converted to NO₂.

Authors' Response to Specific Comment 11

Equation 3 of the AMTD paper demonstrates that NO_x, in addition to NO₂ is detected by the chemiluminescence monitor. This approach has been previously validated, as discussed in lines 1 through 9 of page 835. The statement, "The OMI subscript refers to the average concentration during 12:00–14:00." on lines 20 -21 was a typographical error, and has been removed.

Specific Comment 12

Page 835, line 3: Previous studies suggest that NO_x interference show strong seasonal variation. How did that interference change in winter and summer months at both altitudes?

Authors' Response to Specific Comment 12

This seasonality was accounted for. To clarify this, the authors have added the following discussion to Section 3.1

"The NO_x interference varied seasonally, and had a greater impact on CL NO₂ measurements in the summer versus in the winter. During the months of December – March ($n = 222$; $T = 0.2 \pm 0.9^\circ\text{C}$), the NO_x correction reduced the CL NO₂ concentration at DT by $4 \pm 1\%$ and at CN by $5 \pm 1\%$, while during the months of May – August ($n = 197$; $T = 22.4 \pm 0.7^\circ\text{C}$), the NO_x correction reduced the CL NO₂ concentration at DT by $11 \pm 1\%$ and at CN by $21 \pm 1\%$. The larger NO_x interference correction at the higher elevation during the summer suggests that a greater concentration of reactive nitrogen species is present, as Dunlea et al. (2007) demonstrated that the positive and linear relationship with [NO_x] and [O₃] is due to the photochemical production of reactive nitrogen species alongside O₃."

Specific Comment 13

MAX-DOAS: As stated earlier, shorten this. I do not see much value by giving some terminologies such as “jscript”, “DOASIS”, and “McArtim”. If needed, they should be cited. Please, provide the fitting window for MAX-DOAS retrieval.

Authors' Response to Specific Comment 13

Please see the authors' response to the general comment 5 about the manuscript. The fitting window for the NO₂ retrieval has been included in the Section 2.3 of the manuscript text, as shown below:

“The NO₂ ΔSCD was derived using the fitting window of 405 – 431 nm.”

Specific Comment 14

Page 842, lines 14-18: “The larger NO_z interference correction at the ...” Isn't this sentence telling the same thing as in preceding sentence?

Authors' Response to Specific Comment 14

The authors have removed the reference to Dunlea et al. (2007) in lines 14 – 18, and have incorporated this reference in the discussion provided in response to the reviewer's Specific Comment 12.

Specific Comment 15

Page 845, lines 5-8: This is a subjective statement. The observed difference is not necessarily due to differences in the geographic footprint surveyed. The demonstration is not sufficient enough to claim this.

Authors' Response to Specific Comment 15

The authors have revised this statement as follows:

“The difference between the MAX-DOAS versus in situ-derived NO₂ VCD indicated, but did not prove conclusively, that differences in the geographic footprint surveyed by each instrument impacted the results.”

Specific Comment 16

Page 847, line 20: Replace "retrieved" by "downloaded".

Authors' Response to Specific Comment 16

The authors have maintained the word “retrieved”, since the data was first downloaded and then processed to maintain OMI pixels that met criteria in Table 5 of the AMTD paper.

Specific Comment 17

Table 4 is unnecessary when the information on fitting window and fitting parameters are included in discussions.

Authors' Response to Specific Comment 17

Table 4 has been removed, and the NO₂ fitting window has been included in the manuscript text.

Specific Comment 18

Table 6: NO₂ DT decreased by 8% and NO₂ CN decreased by 12% after applying NO_z interference correction. Why the change in VCD is less than the change in NO₂? Shouldn't it be between 8% and 12%?

Authors' Response to Specific Comment 18

After applying the NO_z interference correction, NO₂ DT decreased by $8 \pm 1\%$, NO₂ CN decreased by $12\% \pm 1\%$, and NO₂ VCD decreased by $7 \pm 1\%$. The reported uncertainty for the average relative difference (%) refers to the 95% confidence interval. Therefore, when the confidence interval is considered, the change in the NO₂ VCD as a result of this correction is not significantly different from 8%.

Specific Comment 19

I think, Figure 3a and b can be combined by using different colors and/or symbols.

Authors' Response to Specific Comment 19

Please see Figure 5 of the revised manuscript provided in the authors' response to General Comment 3.

References for Authors Response to Anonymous Referee #2

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