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Atmospheric Measurement Techniques Discussions

Interactive comment on "Impact of NO_2 horizontal heterogeneity on tropospheric NO_2 vertical columns retrieved from satellite, multi-axis differential optical absorption spectroscopy, and in situ measurements" *by* D. Mendolia et al.

Anonymous Referee #1

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Scientific Significance: Comment 1/1

This paper presents a comparison of three methods to determine tropospheric NO2 columns in the Toronto area (Canada), namely based on satellite, MAX-DOAS and in situ observations. What makes this study unique is the use of in situ observations from the CN-tower in Toronto (instrument at 0.45km above the surface). The manuscript addresses a relevant topic since it studies the link between satellite tropospheric column observations of reasonably large areas and air pollution (NO2) at the surface in a densely populated region. A strong aspect of this manuscript is the literature study,

which gives a good overview of current NO2 monitoring techniques and published comparison results about comparison of satellite and surface in-situ measurements and/or MAX-DOAS. The analysis of MAX-DOAS measurements is straightforward and does not rely on an inversion based on radiative transfer simulations, such as the majority of MAX-DOAS retrieval approaches presented in the last 5-10 years (see the comments below). The most innovative aspect of this work is the method presented to derive tropospheric NO2 columns from observations at the surface and at the tower. However, to this referee, this approach brings up many questions which need to be addressed in more detail.

In the view of this referee, the conclusions presented in Section 4 are not really substantial in the sense that they do not provide new insights in terms of scientific knowledge or major methodological advances with respect to any of the measurement techniques being used.

Scientific Quality: Comment 1/2

With respect to the question if the methods applied are valid, I think it is quite striking that few, or no arguments are given in support of the central assumption for the tropospheric NO2 column derived from the in-situ monitors, namely that it is reasonable to expect a vertical NO2 profile which can be described by an exponentially decreasing function (characterized by a certain scale height and an integrated column amount of NO2), which can be properly constrained by measurements at two fixed altitudes. In my view, this assumption is of such importance for this manuscript that it should at least be accompanied by an elaborate discussion on the validity. For such a discussion, please use the following questions as a guideline:

[A] Please describe why the authors choose an exponential profile shape. The atmospheric pressure profile may show an exponential decrease with altitude, and so may well-mixed long lived species, but why would this be expected for a photochemically active species like NO2? The exponential profile shape may be similar to the 'average'

NO2 profile, averaged over many days/weeks/months, but this does not necessarily imply that this assumption is reasonable for individual cases. In fact, it is probably more likely that realistic profiles show a sharp decrease at some altitude. This sharp decrease may be very low in the morning.

[B] Is the result that most characteristic heights reported in Fig. 2 correspond roughly to the altitude above the surface of the upper in-situ monitor really plausible? Would higher characteristic heights have been found if the upper monitor would have been twice as high above the surface? Why (not)?

[C] Can the authors convincingly show that the reported characteristic heights and tropospheric columns would be substantially different and less plausible if instead of the actual observations of the upper monitor, a single climatological background value would have been used? Or to put it different: can the authors provide argumentation which convinces the reader that the relative contribution of the upper monitor to the retrieved vertical columns and characteristic heights is substantial?

[D] Is criterion 6 (Table 3) in an indirect sense not merely a way to 'ignore' cases that do not correspond to the exponential profile shape? (see also p. 835, l. 16-19). This question is especially important in the case of relatively high (summer noon) atmospheric mixing layers (>0.45km)? Is it not the selection method itself that causes the characteristic heights reported for the summer months to be on the low side, rather than the hypothesis mentioned in the manuscript that the higher photochemical conversion rate in summer leads to lower profiles and lower columns? Although this effect will certainly play a role, the argument would be stronger if it was supported by estimates of the typical vertical velocity in a convective boundary layer, combined with estimates of the NO2 lifetime in this season.

[E] How different would Fig.2 be if only characteristic heights between (for example) 12AM and 2PM local time are considered. This is around the overpass time of OMI, and in addition around the time when the mixing layer is expected to be on its highest.

[F] Would the authors adopt the same approach for other gases, such as ozone? (if so, then see [*] below; if not, then please argue why this approach can be used for NO2 and not for ozone)

[G] How should, with this approach, uncertainty estimates be derived for the characteristic height and tropospheric column? Please comment.

[H] What is the correlation between the tropospheric column and the characteristic height, and how is this correlation interpreted?

[I] Do the retrieved characteristic heights show a diurnal cycle? This diurnal cycles is to be expected to show at least some similarity to the typical diurnal increase of the boundary layer height between the morning and the first half of the afternoon.

[*] If so, the validity of the exponential profile shape assumption could be checked at least for ozone, namely by using a large number of ozone profiles from radio sondes. Using this data set, one could 'simulate' two monitors (one on the ground, and one at 0.45km), run the exponential profile fitting approach, and then test how well the retrieved characteristic heights and tropospheric columns agree with the values for the sonde (only part of the sonde profile should then be taken into account, e.g. only the column below, $\sim$1.0 or 2.0km). Do the retrieved heights show any correlation to the actual profiles? It would be more convincing if the authors could obtain a data set of measured tropospheric NO2 profiles, which covers a wide enough range of profile shapes.

Scientific Quality: Comment 2/2

The retrieval of tropospheric NO2 columns from the differential slant NO2 columns measured with MAX-DOAS is based on geometrically determined air mass factors. This approach is defensible for high elevations (<20 degrees) where it can give a reasonable first order estimate, but questionable for an elevation of 10 degrees, which is also used. The authors do not explicitly mention if they use both elevations to derive the vertical NO2 column from the MAX-DOAS observations, or if they use the 10 degrees observation only for the selection criterion mentioned on p.839 l.15. Also this selection criterion, which leads to the rejection of two thirds of all MAX-DOAS observations is questionable. In a region not far away from sources (i.e. a region where horizontal gradients can be expected) the differences between the differential slant column at 10 degrees and 20 degrees (or 30 degrees) does not necessarily follow the 'rule of thumb' that the differential slant column at 10 degrees is within 15% of the differential slant column at 20(30) degrees. Furthermore, the horizontal gradients do not only lead to observations of different air masses, but also to changes in time, even if the elevation remains unchanged. Are these changes within 15%?

Presentation Quality

The use of the English language is very good, and also the manuscript is well-structured. I think the manuscript could be improved by adding a few more figures, or by describing the content of the figures suggested below in the text.

(A) one Figure showing a map with the city of Toronto, the location of the various measurement sites and (if possible), one example of a small and one of large OMI pixel.

(B) one Figure showing a frequency histograms of tropospheric columns measured with the ground-based/MAXDOAS/OMI instruments.

(C) one Figure showing a frequency histogram of NO2 volume mixing ratios for the low and the high in-situ monitor, both with and without applying the selection criteria (left panel) and a frequency histogram for the ratio of the two, both with and without applying the selection criteria (right panel).

(D) one Figure showing frequency histogram of the characteristic heights

(E) one Figure showing the monthly averaged NO2 volume mixing ratio (similar to Figures 2 and 3) for both stations (both with and without applying the selection criteria).

(F) one Figure showing for at least two example days the diurnal evolution of all relevant parameters: NO2 volume mixing ratio of both in-situ monitors, the characteristic height and the vertical column. If possible, then select one day which shows a typical winter time behaviour, and one day with a typical summer time behaviour. Does the retrieval give a consistent picture which can be understood in terms of NO2 production at the surface, vertical transport and photo-dissociation?

(G) one Figure showing a scatter-plot of tropospheric NO2 column versus characteristic height.

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 825, 2013.