

We thank the referee for carefully reading our manuscript and for giving valuable comments. In this reply, we address all of the referee's comments. The detailed responses are given below (in blue).

General Comments:

This paper presented parameterization method to convert slant column densities into near-surfaced-boxed averaged volume mixing ratios. Retrieved NO₂ mixing ratios were retrieved from this method and long-path DOAS measurements made during MCMA-2006. While, the idea behind this paper using a simple parameterization method is useful and novel, in its current form it is difficult to understand and evaluate. My first concern is as this is a parameterization method the assumptions used should be clearly the many of the assumptions are not clearly stated or poorly presented.

We followed the suggestion of the referee, and revised the paper in order to make the assumptions clearer in Section 2.

Second, the statement that the method does not need vertical profile information is not sufficiently justified as the correction factors are dependent on vertical profile and the authors did not show sufficient evidence that under the assumptions these dependencies disappear. I am, also, unsure of the general applicability of the method and in some places the authors seem to indicate a versatility that does not seem to be justified in this work.

We agree in terms of the dependency towards the vertical trace gas profile. Thus, we introduced the additional prerequisite that also the trace gas dSCDs of the lowest elevation angles must collapse. In this case, the trace gas profile can be approximated as a box profile.

Finally, the overall structure of the paper needs revision as it is hard to follow and seems to jump around quite a bit.

We revised the structure of the paper.

Specific Comments:

Specifically addressing the authors conclusions

Conclusion 2 : "It does not depend on knowing the actual aerosol profile as it is typically necessary for MAX-DOAS concentration retrievals, and can be applied already under conditions of moderately low aerosol load"

I believe that Figure 3 is meant to show that for the lowest angles and for high AOD the correction factor is relatively stable. These plots were hard to understand and did not focus on the information you wish to present. From these plots I see that the correction factor IS profile and angle dependent. I would suggest that rather than presenting all the data (particularly for angle you do not use and AOD's not used) the focus is on the regime within the assumptions so that the reader can see that the correction factor is really within the 10% DOAS error. Also, I think this needs to be coupled with the assumption of the trace gas species vertical profile. In the bottom set of plot the calculation is run for 2 types of trace gases profiles and they appear significantly different. I would suggest that you first show that the O₄ correction factors collapse into a single values for a the regime of angles and AODs appropriate for the model. And then show that these collapsed correction factors are independent of trace gases vertical profile. See comment on conclusion 4

We agree, that the correction factor is profile dependent and rephrased the statement to: "It does not depend on knowledge about aerosols, as it is typically necessary for MAX-DOAS concentration retrievals based on optimal estimation of other inverse methods."

Furthermore, in order to reduce possible confusion we split the figure and only show one case of the upper level to demonstrate the plateau behavior towards the AOD. In a second figure, the lower panel is shown for comparison reasons of wavelength and layer height. The original plots are shown in a supplementary file. See also response to referee 1.

Conclusion3 : “The approach does not suffer from the limited sensitivity of MAX-DOAS at higher altitudes that poses limitations to the use of optimal estimation approaches to infer vertical profiles in situations of high PBL. Yet, this method also can be used as input parameter for more complex retrievals, such as optimal estimation, especially since it provides a reasonably high near-surface vertical resolution (depending on the elevation angle).”

Conclusion 3 seems to be comparing apples to oranges. This is a method for near surface retrievals and does not provide any information about higher altitudes.

We agree, that the language was confusing, and have changed the language as follows: “The approach focuses on those altitudes where MAX-DOAS is maximally sensitive and least limited in information content, which is close to instrument altitude. This near-surface concentration can serve as an anchor for the a-priori estimate in the lowest layer, although it does not provide independent information in the optimal estimation retrieval.”

Conclusion 4: “It does not require a-priori assumptions about trace gas vertical distributions.” I am still unclear on how this method does not make assumptions about the vertical distribution of the trace gas species. I see that for your data you assumed either a box profile or a linearly decreasing profile with height for NO₂. You then apply a correction factor which is profile, angle, wavelength etc. dependent. As stated above from figure 3 I see profile dependence. To clearly demonstrate that the correction factor does not depend on the vertical profile of trace gas species plots of correction factors with differing structures such as: a) A box profile, b) A linearly decreasing layer with height, c) A linearly increasing layer with height, d) A layer slightly aloft should be shown.

We agree, and have modified the text to read: “It only needs some information about the trace gas layer height to apply this method. If the trace gas dSCDs of the lowest elevation angles collapse within DOAS fit error (i.e. have about the same value) the trace gas profile can be approximated reasonably as a box profile.”

Second, we removed the trace gas profile shape discussion from the manuscript. Instead, we introduced the additional prerequisite that also the trace gas dSCDs of the lowest elevation angles must collapse. In case of collapsing the trace gas profile can be approximated reasonably as a box profile.

Figure 4 shows clearly that the correction factor depended on the assumed height of the trace gas layer this is in conflict with the statement of the authors that the method is independent of information on the vertical profile.

Yes, but the correction factor increases with the layer height on a diminishing scale (not linearly), which can be seen in the bottom panel of Fig. 3 or even more clearly in Fig. 4 (dotted lines): an increase of the layer height from 0.5km to 3km (factor 6) in this example leads to an increase of the correction factor from 0.46 to 0.91 (20° SZA, factor 2.0), from 0.59 to 0.97 (50° SZA, factor 1.6) and from 0.31 to 0.83 (80° SZA, factor 2.7). Furthermore, with increasing layer height the correction factor becomes less sensitive towards changes of layer height. For layer heights at or above one km the correction factor vary typically less than 30% for doubling or halving of the layer height (for <60° SZA). Also, with longer wavelength the sensitivity towards layer height decreases.

1) The discussion of the correction factors need expansion and restructuring as the method and the authors conclusions depend on the correction factors in a specific range being independent of the trace gas vertical profile more time is needed on demonstration this fact. The discussion of the correction factor on Page 13 line 3 needs to be in the method section.

We removed the discussion about the trace gas vertical profile (see above) and restructured the method section (including the discussion on p. 13).

2) The description of the method and explanation of figure 1 need reworked. It is cumbersome to read and understand as it jumps from what is needed for your collapsed dSCD and a traditional ground based DOAS method. I would suggest a comparison between what occurs in the higher angle (traditional ground based DOAS retrieval) and the lower layers (your collapsed method). On page 5 line 29 you could add the contrast to the higher elevation and then go on to clearly state your assumptions a) Strongly attenuated light path (High AOD verified by collapse of the lowest elevation angles into a single light path L_{eff}) b) L_{eff} must terminated within the trace gas layer c) I do not see the added value of the scattering probability panel, it adds un-needed complexity and interrupts the flow of the text.

The description of the method and Figure 1 was revised following mainly the referee's suggestions. We think that the scattering probability panel is useful since it emphasizes that in reality a measurement does not consist of just one single scattering event but is composed of many scattering events overlaying each other and following the scattering probability graph. We added this statement into the text.

3) There is no discussion about decreased sensitivity. As the light path is attenuated the path length is shorter in the troposphere what effect does this have on detection limits?

We cannot see that this approach comes along with a detection limit issue. The approach systematically exploits only the information in the lower elevation angles. Amongst other factors the amount of light reaching the detector determines the detection limit. This amount is often even higher in the lower elevation angles due to aerosols. Aerosols also shorten the effective path length, but regardless, the lower elevation angles are most sensitive (largest sensitivity of MAX-DOAS is near the instrument altitude).

Technical Comments: Page 1 Line 30: Is the agreement with well mixed layers evidence that there is more of dependence on the vertical profile than assumed as you assumed a well mixed profile?

We are not sure we understand the referee's comment. There is no circular argument in our logic. Under the criterion of collapsing NO_2 dSCDs, the approach implies that the observed trace gas layer can indeed be approximated as box profile. The existence of vertical gradients follows from comparison with LP-DOAS near the surface. These gradients are below the line of sight of the MAX-DOAS instrument, i.e., near the roof-line few meters above street canyons, which were probed by LP-DOAS. We do not see a contradiction to our assumptions.

Page 4 Line 25: Remove the "Then"

Done.

Page 5 Line 17: Remove the "Then"

Done.

Page 5 Line 22: "it" change to "sunlight"

Done.

Page 5 Line32 to Page6 line2: This sentences is unclear as what plays a minor role

This sentence was removed by revising the paragraph describing Fig. 1.

Page 6 Line 2: Remove the “Then”

Done.

Page 6 Line 5 and 6: not sure what you try to say by “cancels out applying the DOAS method...” I do not think you can cancel a method, under certain conditions the advantages of making DOAS observations on multiple angles may not yield information about the vertical profile but it does not cancel the method.

It is not stated that a method is canceled (out). In order to make the sentence clearer it has been changed to: “The light path length from the sun to the effective height of the scattering events of a low elevation angle is almost as long as the one for a reference spectrum to the same height (especially if the reference is acquired close in time to the measurement spectrum). Thus, the according absorption signals (covering the distance from the sun to the effective height of the scattering events) in the two spectra mainly cancel out applying the DOAS method resulting in differential effective path lengths.”

Page 6 Line 13: the light is less constrained is awkward, think about stating that is is a place where your assumption about L_{eff} breaks down.

This sentence was removed by revising the paragraph describing Fig. 1.

Page 8 Line 5: shouldn't equation 3 be sin not tan?

This is correct. The calculation of h_{eff} was made with the right equation though (using sin).

Page 8 Line 7: Figure 2 either needs to be cut as it added little or expanded to include the equations and assumptions used as is it adds little that the equation on their own do not

We respectfully disagree with the referee. The diagram gives a quick overview of the one-step approach which we consider to be useful.

Page 9 Line 19: Figure 3 this figure does not illustrate the collapsing correction factors so I would re-work see comment above but independent of that. the figure is unclear is what vertical profiles are used for the top row, if all box profile why? For bottom row what is the elevation angle. Also use same y scale for all plots as it will help in clarity

We split and changed this figure (see above). The information about the elevation angle (3°) can be found in the manuscript. We added it into the figure caption.

Page 10 Line 13: Poorly worded sentence

We removed this sentence.

Page 10 Line 17: “constantly decreasing profile” do you mean “linearly decreasing” or something else?

The linearly decreasing profile is now discussed in the Supplementary Information section.

Page 10 Line 18: Did not understand this sentence are you stating that a linearly decreasing profile would result in the divergence of L_{eff} sooner

Yes, but this does not apply any longer with the additional condition for collapsing NO_2 dSCDs.

Page 10 Line 26: triangle profile should be linearly decreasing profile

See above.

Page 11 Line 14: The discussion of a collocated layer and fixed trace gas layer is confusing

We changed it and hope it is clearer now.

Page 11 Line 20: Figure 4 I think the x title should just be aerosol layer height the height of the NO₂ layer is in the legend, I first thought it was a ratio between the layer heights.

We agree and changed it accordingly.

Page 20 Line 1: I find this section difficult to follow as higher and lower are not good references. I would rework the figure to have circles and triangle and square to refer to each type of points

We followed the suggestion of the referee.

Page 21 line 16 solar relative azimuth angle (SRAA) should be relative solar azimuth angle (RSAA)

We changed “SRAA” to “RSAA” throughout the manuscript.