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

The manuscript entitled 'Parameterizing radiative transfer to convert MAX-DOAS dSCDs into near-surface box averaged mixing ratios and vertical profiles' by Sinreich et al. describes a new method for the retrieval of near-surface trace gas concentrations from MAX-DOAS data. Several approaches for the retrieval of aerosol and trace gas information from MAX-DOAS measurements exist in the literature, which are usually based on a two-step procedure, during which in a first step aerosol profiles are retrieved, and in a second step trace gas profiles. The approach by Sinreich et al. aims for simplifying the retrieval algorithm in a way that a retrieval of the aerosol profile can be avoided and trace gas concentrations can be derived directly from the measurements. This represents a novel approach which, if successful, would yield a simple estimate of the near-surface abundance of trace gases. Therefore the manuscript presents novel ideas and addresses relevant scientific questions within the scope of AMT.

However, the manuscript lacks of a concise presentation of the retrieval methods. Parts of the algorithm are described in the results section rather than the methods section. Several unsupported assumptions are made, for example on the layer height of the NO2 profile. The algorithm only works for relatively high AOD, but it is not described if and how a filtering of the data according to AOD has been performed. Several parameters are varied simultaneously (like wavelength and layer height) in the presented sensitivity studies, making it impossible for the reader to determine which parameter is causing which effect.

We reorganized the paper and moved the mentioned parts to the method section. The layer height is supported by two studies (as is being addressed in the separate referee comment, see below). The description of the filtering has been made clearer. Only, values were considered following the collapsing criteria of O_4 and NO_2. We do not see how the referee could come to the conclusion that wavelength and layer height are varied simultaneously. Fig. 3 lower panel clearly shows the effect if one parameter is changed. However, we have reorganized the Figures to separate effects to maximize clarity.

In contrast to other well established methods (optimal estimation and parameterized retrievals), a particular weakness of this method is that it relies on assumptions on the trace gas profile shape, which is usually not available.

The same weakness applies to parameterized retrievals where also assumptions on the trace gas profile have to be made and, to some extent, to optimal estimation where the a-priori profile information is typically part of the retrieved profile.

A discussion on how to overcome this problem is missing. Furthermore, I disagree with many of the conclusions reached by Sinreich et al. which are cited here in italic :

• *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*

The approach presented here suffers from the same limited vertical resolution as any other retrieval approach. The limited vertical resolution and lack of sensitivity for high altitudes is a result of radiative transfer and the underlying physics, and not of the particular retrieval algorithm. The question how the method of Sinreich et al. competes with other approaches regarding the accuracy of the retrieved surface concentrations has not been addressed.

We agree, and did not mean to imply otherwise. The particular benefit of the approach presented here, is that UNDER CERTAIN ASSUMPTIONS (spelled out clearer in the revised paper) the measured dSCDs can be converted into near surface VMRs WITHOUT knowledge pre-requisite information about aerosol extinction.

The bullet text in the conclusion section now reads: "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."

A comparison of the parameterization approach presented here with Optimal Estimation and Tikhonov Regularization inversion schemes is currently in preparation (Ortega et al., in prep; Comparison of Tikhonov regularization and optimal estimation inversion techniques to retrieve trace gas vertical profiles from MAX-DOAS). These approaches DO NEED pre-requisite information about aerosol extinction.

• *Yet, this method also can be used as input parameter for more complex retrievals, such as optimal estimation...*

Using information on the atmospheric state obtained from the same set of measurements as a priori for optimal estimation algorithms is somewhat circular. A priori information needs to be independent.

Agreed. This is made explicit in the revised bullet text.

• *It does not require a-priori assumptions about trace gas vertical distributions.*

In order to retrieve surface concentrations using the approach presented here, the actual vertical distribution of the trace gas layer needs to be known since it strongly affects the correction factor. This results in much higher uncertainties than for other algorithms which aim to retrieve the profile shape, either in parameterised form or as a discrete vertical profile.

We agree partly with the referee's comment. We acknowledge that the trace gas profile shape is unknown. We have revised the manuscript to include a condition for collapsing NO_2 dSCDs in the lower elevation angles. With this constraint, the profile shape is no longer an unconstrained uncertainty, and can be approximated reasonably as a box profile. But even without this constraint, we further think that the uncertainty due to the uncertain profile shape is not higher than for other algorithms, but in fact it is lower. This is because the correction factor to derive the near surface VMR does not scale linearly with trace gas layer height, but the sensitivity is buffered, and the correction factor approaches unity for high layer heights. This is also shown in Figs. 3 and 4.

The revised bullet text reads: "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."

• *It is applicable in cases of limited spectral coverage of the spectrometer (only one O4 absorption band).*

As far as I know, this applies to all currently existing aerosol retrieval algorithms.

We agree. That's why in the paper we explained this statement with the sentence the referee quoted as next point. See also our response to following comment.

• *Optimal estimation and other retrievals in principle can use only one O4 band, but face limitations in air masses with high PBL such as Mexico City, where aerosol inferences from*

use of only one O4 absorption band can yield unstable results, but remain pre-requisite to derive trace gas information.

I cannot see why the presented approach should be more sensitive for high PBL than other retrieval algorithms. The sensitivity for higher altitudes (or its lack) is a question of radiative transfer and viewing geometry. It has been clearly demonstrated by Clemer et al. (2010) that aerosol profiles can be reliably retrieved from O4 measurements at any absorption band. Unstable results can be avoided using appropriate a priori constraints.

In Clemer et al. (2010) the discussed aerosol scenarios are limited to cases with an aerosol scale height of around 1 km, which is below the altitude where averaging kernels show still good sensitivity (up to 1.3 km at 360 nm [Sinreich, 2008]). Already under these conditions the mean degree of freedom of signal decreases from 2.14 to 1.79 from 577 to 360nm. There are no cases for higher aerosol layer heights discussed in Clemer et al., 2010 as it would apply to Mexico City (up to 2-4km high PBL). As soon as the aerosol scale height exceeds the altitude where averaging kernels indicate no longer good sensitivity, the O_4 approach does not provide a stable aerosol profile. Based on our own attempts of optimal estimation inversion in Mexico City based on O_4 retrievals at 360nm we were unable to obtain stable results [Sinreich 2008].

The approach presented here does not run into this limitation. Rather it benefits from conditions of high aerosol optical depth, and aerosol layer height. Under such conditions, the correction factor for the lowest elevation angles approaches unity (no correction needed) and becomes less sensitive to profile shapes. In contrast to other approaches this method systematically only exploits the information content that MAX-DOAS derives from the lowest layers of the PBL.

We added in this bullet: "As soon as the aerosol scale height exceeds the altitude where averaging kernels indicate no longer good sensitivity (about 1.3km at 360nm), the O_4 approach does not provide a stable aerosol profile (Sinreich, 2008)."

• *Especially if the spectral range does not include the the 477nm O4 band or longer wavelengths the information content for aerosol retrievals is relatively limited due to strongly increased scattering at shorter wavelengths.*

Again, the approach presented here suffers from the same limitations with respect to the information content as the other approaches. The difference is that, in contrast to optimal estimation, this approach does not allow to quantify these limitations. Clemer et al. (2010) have demonstrated that the information content is not much smaller at 360 nm than at 477 nm, in particular in polluted environments where the algorithm presented here is applicable only.

See our previous answer.

Specific comments

The basic idea of the method described by Sinreich et al. is to use the observed O4 dSCD as a proxy for the light path, which then allows for deriving near-surface concentrations of NO2 form NO2 dSCDs without explicit knowledge of the aerosol profile, and without using a complicated retrieval algorithm. A direct conversion of the NO2 dSCD to a concentration would be possible if the shape of NO2 and O4 profile would be equal. To account for the difference in profile shape, a correction factor has been introduced which accounts for the differences in path length (or airmass factor) for O4 and NO2. This correction factor, determined using radiative transfer modelling, is a function of AOD, aerosol layer height (do be precise also aerosol profile shape), as well as NO2 layer height and profile shape. The method is only valid for relatively high AOD (> 0.3) and small elevation angles (< 3°), and therefore restricted to polluted environments such as Mexico City.

We agree partially. The correction factor is not a function of AOD, but rather it becomes independent of AOD (as long as the aerosol load is high enough that the O_4 dSCDs collapse). The figures showing the correction factors versus AOD is an intermediate step in order to determine the (AOD independent) correction factor. We have changed the text in the methods section in order to make this clearer.

While the dependence on aerosols can be eliminated by this method to some extent, there is still a very strong (almost linear) dependence of the correction factor on the layer height and profile shape of NO2, see bottom panels of Fig. 3.

This is not correct. 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.

There is no information on how this layer height can be derived or how it can be dealt with this lack of information. Instead, a NO2 layer height has simply been assumed for the MCMA measurements without further comment where this information comes from. How can reliable surface concentrations be determined if the NO2 layer height and profile shape remain as unknown parameters?

This is not correct. The choice of the layer height in this paper is not arbitrary. As mentioned in the text, it is in agreement with measurements of the PBL made by de Foy et al. (2005) in MCMA-2003 and by K. Knupp and D. Phillips from The National Space Science & Technology Center of the University of Alabama at Huntsville during MCMA-2006. We agree with the referee in terms of the trace gas layer shape and introduced a prerequisite to constrain the layer shape, which consists in collapsing NO_2 dSCDs. Under these assumptions (spelled out in Section 2 of the paper) the trace gas profile can be approximated reasonably as a box profile.

7647.21 It is mentioned that a 'collapsing' of the O4 dSCDs at high AOD is a necessary prerequisite for the validity of the method, because the scattering events than happen at comparable distances. Please explain why this needs to be the case. To me it appears that instead a correction factor that does not vary much with AOD is a necessary prerequisite, and that the 'collapsing' is rather an indicator for a high AOD where this prerequisite is fulfilled.

This is a matter of semantics. The accessible quantity available to define a criterion is the O_4 dSCD. Hence our criterion is based on the collapsing O_4 dSCD. Actually as we had written in the sentence that the referee is referring to, the collapsing of the O_4 dSCDs is not a prerequisite for the validity of the method, but for applying the method. Indeed it is an indicator for a high AOD.

Equation 3 is wrong. It must be

$h_{eff} = dL_{eff} \cdot \sin \alpha$.

We agree. The calculation of h_{eff} was made with the right equation though (using sin).

Fig.2: I suggest to remove this complicated diagram and to show the two simple equations it represents in the text. I furthermore suggest to move Equation 4 and the corresponding explanation (7652.26 ff), which describes how the correction factor is actually calculated,

from Section 3 to Section 2 since this Equation is an essential part of the method. I do not understand what $c_{retrieved}$ and c_{real} mean in the context of Equation

We agree partially with the referee. The diagram gives a quick overview of the one-step approach which we consider to be useful. It is not too complicated. We moved Equation 4 into Section 2. $c_{retrieved}$ is the concentration which is retrieved by radiative transfer calculations if $L_{eq,O4}$ is used as path length. c_{real} is the concentration which is input for the radiative calculations which is considered to be the real concentration. We added this in the text.

4. I was able to reproduce how you derived the right side of Equation 4, but this is not directly evident and should be explained in some detail. The radiative transfer model (McArtim) should be introduced in section 2, where the correction factors calculated with McArtim are discussed (McArtim is first mentioned in section 3.1).

We explained Equation 4 in more detail and introduced McArtim already in Section2.

It is not described how exactly differential slant column densities are measured/modelled. Did you use a fixed noon reference or a zenith sky reference close to the off-axis measurement? These different approaches should yield a very different diurnal variation of the correction factor.

We used reference close to the off-axis measurement. For the radiative transfer modeling, we used a reference with the same solar zenith angle. We added this information in the manuscript.

7649.2: It is mentioned that the NO2 profile has a constant mixing ratio within the PBL, but the shape of the aerosol profile is not specified. Are aerosol layer height and PBL height equal?

Yes, they are equal. We added this in the manuscript.

Upper panels of Fig. 3: The selection of different parameters for the calculation of the correction factor is confusing. In each of the upper panels, not only the wavelength but also the layer height is varied. This makes it impossible to judge whether the changes are caused by the choice of the wavelength or by the different layer heights. Please only vary one parameter at once. My suggestion is to show only two wavelengths (e.g., 360 and 477 nm), and to show for each wavelength the results for two different layer heights.

We respectfully disagree, as the effects of different layer heights and wavelengths can be seen clearly in the lower panel. Nevertheless, we have separated the information contained in Fig. 3 into two separate Figures in the revised manuscript. One Figure now only shows a single case of the upper panels (of former Fig. 3) to demonstrate the plateau behavior towards the AOD. In a second figure, the lower panels are shown for comparison reasons of wavelength and layer height (NO_2 box profiles only). The original plots have been moved in a supplementary file, as they broaden the results towards a linearly decreasing NO_2 profile shape, and additional wavelengths.

7650.9: Different NO2 profile shapes and altitudes result in tremendous changes in the correction factors (see lower panel of Fig. 3), and that also the range of AOD values where the algorithm is valid is also a function of the profile shape. Again, how do you know the profile shape beforehand?

See above.

7650.14: I do not understand this sentence. What do you mean with 'regular MAX-DOAS splitting' and in what respect is a triangular profile shape producing an additional effect?

Again, how can you distinguish between triangular and box-shaped profiles (or any other profile shape)?

We removed the discussion of different trace gas profile shapes from the manuscript, based on the additional criterion of collapsing NO_2 dSCDs. In fact, the addition of this criterion did not change the dataset compared to our initial submission. The triangular profile shape is now discussed in the Supplementary Information file. Then, NO_2 dSCDs not necessarily collapse, or collapse at higher AOD.

7651.7ff: The discussion in this paragraph, and the data shown in fig. 4, again show that the method described here cannot be applied without independent knowledge on the trace gas profile shape. For the data in fig. 4, it has not been mentioned if the AOD or the extinction were kept constant while the aerosol layer height has been varied.

We agree, and have removed profile information from parameterization of MAX-DOAS data from the manuscript. The only profile information now comes from comparing with ground-based LP-DOAS (Fig. 9).

7652.15: It is stated that AOD values between 0.3 and 0.6 would reflect the situation during the MCMA-2006 campaign. However, from the sun photometer measurements at site T0 available via Aeronet, the 380 nm AOD was frequently above 0.6, and often reached values of more than 1 (and even 1.5 on March 24). I wonder why it is speculated here about the aerosol load from the qualitative behaviour of the MAX-DOAS data if AOD measurements are readily available. Furthermore, it would be very instructive to see some days of O4 and NO2 dSCDs (e.g., the three days from Fig. 6), in order to be able to reproduce what is stated here regarding the qualitative behaviour of the MAX-DOAS data.

We have added the O4 and NO2 dSCDs for three chosen days of LP-DOAS comparison. The crucial AOD value is the lower value since it is the start of a plateau in the correction factors which means that at higher AOD the correction factor remain (nearly) constant. The upper AOD value is only used for confinement reasons and to reduce noise in the RTM with higher AODs. The results don't depend on this number. The revised text reads:

"Also, at times overlapping of higher elevation angles could be observed for both O_4 and NO_2 indicating a higher AOD. However, due to the plateau formation, a higher AOD typically has only a small effect on the correction factor."

7652.26ff: As already mentioned above, I suggest to move this paragraph to section 2 since it describes the general method.

Done.

7652.12ff: Here an arbitrary diurnal variation of the boundary layer height has been chosen without further motivation. The Foy et al. 2005 article is missing in the reference list…

Actually de Foy et al. (2005) can be found in the alphabetically ordered reference list at the letter 'd'.

…, and it is not described what kind of measurements Knupp and Phillips performed.

Knupp and Phillips used a radar profiler, which is described in Knupp et al. (2006). We have added an according statement in the revised manuscript.

Knupp, K. R., Walters, J., and Biggerstaff, M.: Doppler Profiler and Radar Observations of Boundary Layer Variability during the Landfall of Tropical Storm Gabrielle. J. Atmos. Sci., 63, 234–251. doi:10.1175/JAS3608.1, 2006.

An accurate estimate of the trace gas layer height as well as profile shape, crucial for the accuracy of the method, is not provided. The correction factor varies almost linearly with

trace gas layer height and also with profile shape. These important parameters are likely to vary from day to day and need to be known for the time of each individual measurement. Furthermore, the height of the NO2 layer is usually decoupled from the boundary layer height (e.g. Wagner et al., 2011), whereas it appears that NO2 and aerosol layer heights are assumed to be equal.

See above. The correction factor does not vary closely linear to the layer height, and is thus less crucial to know then the referee makes it appear. As mentioned above, we assume the aerosol layer and trace gas layer to be collocated, but we have also explored the sensitivity in case they are not collocated (see initial Fig. 4). We agree that it is easily possible that the trace gas layer is decoupled from the aerosol layer. In such cases, usually the aerosol layer is higher than the NO_2 layer (due to shorter life time). Fig. 4 demonstrates that this has a minor effect on the correction factor.

7654.6ff: Again, two parameters are varied simultaneously, namely the relative solar azimuth angle and the boundary layer height. Therefore it is not possible to decide to what extent the variation of the correction factor shown in Fig. 5 depends on SRAA on the one hand and on BL height on the other hand. Furthermore, the quantitative statements made on the SRAA dependence are difficult to reproduce because the SRAA is not plotted in Fig. 5.

We respectfully disagree with the referee. The purpose of Fig. 5 is to show the correction factors of the measurement setup in Mexico City in 2006 and the focus of the figure is to demonstrate the relative differences of the RSAA, which can be seen at a given layer height since all 4 directions (East, South, West and North) are shown. Also, the effect of the changing layer is observable in the North and West direction, which hardly are affected by a changing RSAA. However this effect is already demonstrated in Fig. 3.

The point we want to make with this figure is that the correction factor changes significantly when measuring towards the sun, in particular if the PBL is low. Our setup in Mexico City avoided these conditions, but we show calculations for this case in the figure to illustrate this sensitivity. We revised text to make this clearer. While we assume it is common knowledge that the sun rises in the East and travels through South (northern Hemisphere) to the West, we have added a panel to Figure 5 (now Figure 6) that explicitly shows the RSAA for the four directions.

Equation 5: This equation is the mathematical representation of Fig. 2 and should be moved to section 2. The symbol m for the conversion of concentration to mixing ratio could potentially be confused with the symbol for mass.

Done. Symbol ‘m’ was replaced by ‘CF’.

7655.8: To what extent can the difference between MAX-DOAS and LP-DOAS (e.g., in the morning of March 23) be attributed to wrong assumptions regarding the NO2 layer height and profile shape?

We have added the following statement to the text: “During the peak in the morning of March 23, LP-DOAS shows in average about 1.7 times more NO_2 than MAX-DOAS. At the time of the peak (9:35 am), the PBL was assumed to be about 700m. The layer height in this approach would have needed to be around 300m in order to match the NO_2 VMR of LP-DOAS. Thus, we think that this difference is mostly a vertical inhomogeneity.“

7655.19: It is stated that the differences in retrieved VMR for the different viewing directions in the late morning confirm the previously discussed azimuth effect. However, this should be accounted for by using the appropriate correction factors as a function of relative azimuth angle. Is it possible that the differences rather indicate that either the correction factors are (slightly) incorrect or that there are horizontal inhomogeneities?

We think that the referee misunderstood our statement. Actually, it is stated that the compliance of the three directions other than in the late morning indicates a confirmation for the right azimuth consideration. Every direction has its own set of correction factors accounting for the azimuth effect. We changed the sentence to make this clearer, and cannot see how the referee concludes possible incorrect correction factors.

Section 3.3: Here an attempt is made to derive vertical profiles from the measured average concentration from different elevation angles, using a simple geometric approach. First of all, it appears that the method is incorrect because the average concentrations need to be weighted with the respective layer heights rather than just taking the difference between the average concentrations from different elevations.

Unfortunately, the description of the method did not represent what was actually done, and we agree with the referee and added a proper description.

Second, the fact that scattering does not occur at a specific location in the atmosphere, but is rather smeared out along the line of sight, is not considered. A discussion on the validity of this simple approach, as well as on the resulting uncertainties, in particular in comparison to other established profile retrieval algorithms, is missing.

We partially agree. The smearing out is in fact accounted for by introducing an 'effective' scattering height. The scattering events indeed occur at different altitudes around this height, which is reflected by the choice of 'effective'. But we agree that the profile retrieval using different MAX-DOAS elevation angles does not have enough information to derive a proper profile (~ 1 degree of freedom from the near surface VMR). We changed the paper accordingly.

7657.23: Here a general statement on the increase in vertical resolution with increasing number of measurements at different elevations has been made, without providing any numbers. What do you mean with 'relatively high' vertical resolution? From other publications (e.g., Friess et al., 2006), it is well known that the number of independent pieces of information (usually less than 3) is significantly lower than the number of elevation angles and that neither information content nor vertical resolution can be indefinitely extended using additional viewing directions.

We agree and removed this statement.

Technical comments

7654.10: replace 'RSAA' with 'SRAA'

We now consistently use RSAA throughout the manuscript, adopting the suggestion by referee 2.

Equation 1: I suggest to replace c_{avg} with the commonly used symbol $\bar{c}$. It would be useful to mention that the path length dl is equal to the more commonly used box-amf times the layer height.

We changed the symbol. However, we do not see a benefit of introducing the air mass factor in this section of the paper.

7651.8: replace 'altitude' with 'altitudes'.

Done.

7655.18: replace '%-tiles' with 'percentiles'.

Done.

Lower panels of Fig. 3: It is not specified for which AOD and for which elevation angles the values were calculated.

As mentioned in the text these are mean values for AODs between 0.3 and 0.6 for 3° elevation angle. We changed the text to make this clearer.