

Interactive comment on “Parameterizing radiative transfer to convert MAX-DOAS dSCDs into near-surface box averaged mixing ratios and vertical profiles” by R. Sinreich et al.

Anonymous Referee #1

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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

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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.

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

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surface concentrations has not been addressed.

- *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.

- *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.
- *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.

- *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.

- *Especially if the spectral range does not include the the 477nm O4 band or longer*

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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.

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 from 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 air mass 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 ($< 3^\circ$), and therefore restricted to polluted environments such as Mexico City. 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. 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?

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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.

Equation 3 is wrong. It must be $h_{eff} = dL_{eff} \cdot \sin \alpha$.

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 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).

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.

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?

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

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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.

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?

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)?

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.

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.

7652.26ff: As already mentioned above, I suggest to move this paragraph to section 2

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since it describes the general method.

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

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.

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.

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?

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

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correction factors are (slightly) incorrect or that there are horizontal inhomogeneities?

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. 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.

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.

Technical comments

7654.10: replace 'RSAA' with 'SRAA'

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.

7651.8: replace 'altitude' with 'altitudes'.

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

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

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