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Atmospheric Measurement Techniques Discussions

Interactive comment on "Inversion of tropospheric profiles of aerosol extinction and HCHO and NO_2 mixing ratios from MAX-DOAS observations in Milano during the summer of 2003 and comparison with independent data sets" *by* T. Wagner et al.

T. Wagner et al.

thomas.wagner@mpic.de

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General comments This paper presents an inversion scheme retrieving profiles of aerosol extinction and HCHO and NO2 mixing ratios from MAX-DOAS. The inversion scheme is applied to MAX-DOAS observations (with a unique setup using three telescopes) during the FORMAT campaign. The results are compared with independent measurements and intercompared for the different telescopes to assess the ability of the inversion method. Moreover, by additionally developing a cloud classification algorithm, potential effects of clouds on the profile inversion are investigated. An assessment for the ability of the profile inversion performed in this way is unique and the subject of this paper is appropriate for AMT. However, I raise some points that require additional consideration. For example, I am afraid that the presented inversion scheme may be too incomplete to give a systematic interpretation for the inversion results. I recommend this paper will be published after adequately addressing my concerns, which are described in detail below.

Author reply: We thank the reviewer very much for the positive assessment and the recommendations for improvements. Our detailed answers to the specific points are listed below.

-We added more discussion about the stability of the profile inversion results (mostly in section 3.4). We also added new sensitivity studies about the influence of the initial values on the results of the profile inversion (new Fig. S2 in the supplement) It turned out that the retrieved profiles are almost independent on the initial values. Thus we conclude that the dependence on the initial values is not the main problem for unstable inversion results. We conclude that the instabilities are mainly caused by effects, which are not explicitly considered in the forward model (like the influence of clouds or horizontal gradients). Additional instabilities arise from ambiguities (e.g. from elevated layers). Surprisingly even for such cases, our inversion procedure yields meaningful results for most observations. We added this information to the revised version.

-We added new information (new Fig. 4) about the conditions, for which the aerosol retrievals with shape parameter S £ 1 fail. For observations on clear days with layer heights below 1.2 km, the retrieved AOD only slightly depends on the assumed shape parameter.

-We added a comparison of retrieved aerosol extinction between the different telescopes (in Fig. S7 in the supplement (old Fig. S4).

-We added more simulation studies on the effect of elevated aerosol profiles on the profile retrieval with shape parameter S =1 (new Fig. S4 in the supplement). These simulation studies support the findings of Fig. 5 (new Fig. 6).

-We added results of a simulation study on the horizontal sensitivity range of MAX DOAS observations (new Fig. S1 in the supplement).

-please also note (although no major change) that we replaced the term chi square by the correct definition (residual sum of squares, RSS), see new equation 15.

Specific comments

As mentioned at several places in the manuscript, we need to pay much attention to the stability of the retrieval. Because of this, I am afraid that the presented inversion scheme is too incomplete to give a systematic interpretation for the inversion results. For example, the results may change considerably in the retrievals that use different initial values. Since the instability comes from the fact that the problem the inversion method tries to solve is ill-posed, would it be a reasonable way to add the results of AOD and VCD, which are retrieved under stable conditions with prescribed values of both layer height and shape parameter?

Author reply: This is an important point, and we did not pay enough attention to it in the original version. Motivated by the reviewer's suggestion we carried out additional sensitivity studies about the influence of the initial values. It turned out that for most measurements (about 60%) stable inversion results could be obtained even if all three parameters were freely fitted. Our standard inversion performed even better: if the shape parameter was set to a fixed value, over a wide range of initial values almost identical results were obtained for almost all observations. Thus we conclude that the dependence on the initial values is not the main problem for unstable inversion results. We conclude that the instabilities are mainly caused by effects, which are not explicitly

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considered in the forward model (like the influence of clouds or horizontal gradients). Additional instabilities arise from ambiguities (e.g. from elevated layers). We added this information to the revised version.

I think that this study clearly indicate the need for the optimal estimation method, and recommend stating this in the manuscript, although I realize the authors use a different strategy without a priori profile information.

Author reply: As the reviewer might imagine we do not fully agree to this conclusions (see e.g. also the comments of the other reviewer about negative values). We added the following sentences to the introduction: 'It should be noted that both retrieval methods (optimal estimation and the parameterisation approach) have their advantages and disadvantages, and that the importance of these advantages and disadvantages is seen differently by different research groups. In our opinion, a main disadvantage of our approach is that it can not retrieve 'complex' profile shapes like e.g. two layer profiles. One of the main advantages is that it is a very stable and robust method (see below).'

I understand from the presented radiative transfer simulation that the diffusing screen effect can lead to an underestimation of AOD. In reality, however, cloud particles can change the path length in a way similar to aerosol particles, so that a clear separation of AOD from the cloud optical depth (COD) is very difficult with the inversion using O4. In this case, it is expected that the retrieved AOD should contain a contribution of COD, leading to overestimation of AOD. Is it reasonable to add this additional important influence of clouds? Why don't the AOD retrieved in this study show such overestimation?

Author reply: The influence of clouds (or aerosols) on the MAXDOAS observations depends a lot on the altitude. The effect of clouds is particularly complicated, because for MAXDOAS inversions the difference of the O4 absorption for a low elevation angle and for 90° of the same elevation sequence has to be considered. Thus no simple intuitive interpretation is possible, and clouds can lead to both increase or decrease of the re-

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trieved AOD. An increase, however, is only found for very low clouds. We think that this information was already given in the original version of the manuscript. In addition, the retrieved AOD under cloudy conditions supported the radiative transfer simulations. In the revised version we added the following text to section 4.1.1: 'Here it is important to note that optically thin clouds have in general an opposite effect compared to aerosols close to the surface, and the cloud OD will not simply add to the AOD in the aerosol inversion.'

It seems to me that section 3.4 is hard to understand. Please consider to simplify it. As mentioned in section 3.4, the retrieved aerosol layer height requires a correction factor as high as 2. I am afraid that this correction is crucial and it is unfair to say that the inversion method presented in this study can retrieve a vertical profile.

Author reply: We added more information about the pragmatic choice of shape parameter S = 1.1 for the retrieval of AOD at the end of section 3.4. Although we agree that the information of our profile inversion is limited (only AOD and approximate layer height), we want to keep the term 'vertical profile' in our paper, because we think that information about the layer height from passive remote sensing is unique and very useful. We added the following general sentence about the limitation of our profile retrieval at the beginning of section 3: ' It should be noted that the profile information from our retrieval is limited. Besides the integrated quantities (trace gas vertical column density or aerosol optical depth) usually only a characteristic layer height is derived, and one could speculate whether this information is sufficient to characterise a vertical profile. Nevertheless, in our opinion the retrieval of a layer height from passive remote sensing is a unique and very important information. Thus we will use the term profile for the results of the MAX-DOAS inversions presented in this study.'

Technical corrections

Please use a word, FORMAT, FORMAT 2003, or FORMAT-II campaign, consistently throughout the manuscript.

Author reply: We now use FORMAT-II throughout the text.

p.3898, line 25: "formaldehyde" should be "HCHO". "nitrogen dioxide" should be "NO2".

Author reply: corrected

p.3899, line 15: "acuracy" should be "accuracy".

Author reply: corrected

p.3899, line 15: Why is the accuracy for ultra-light research aircraft measurements better than that for ground-based measurements?

Author reply: The instrument on the ultralight aircraft is an upgraded system with a new small size fluorimeter with better temperature stabilization than the commercial instruments used at the ground. The better temperature stabilization results in both, improved precision and accuracy. We added this information to the text.

p.3899, line 27: Why does a full sequence take 10 min, while a single measurement of 90 sec is made five times (the number of elevation angles) in the sequence?

Author reply: The additional time results from the motor movements. We added this information to the text.

p.3900, line 8: Please explain why Fraunhofer reference spectrum is better to contain small atmospheric absorptions.

Author reply: In principle also Fraunhofer reference spectra with large atmospheric could be used, because the respective offsets in the DSCDs would cancel by calculating the difference between the DSCDs for low elevation angles and zenith measurement for individual elevation sequences. We removed this statement from the text.

p.3901, line 9: While the O4 VCD varies with temperature and pressure, a constant O4 VCD has been assumed. How much is the actual diurnal variation of O4 VCD due to

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changes in temperature and pressure? Does it influence the accuracy in determination of AMF?

Author reply: The influence of changing air pressure on the O4 VCD is below 2% and can be neglected. Also the effect of changing temperature is expected to be negligible, but more difficult to be quantified, because of the rather large uncertainty of the temperature dependence of the O4 cross section (see e.g. Wagner et al. 2002). We added this information to the text.

p.3903, line 4: It is stated that the trace gas concentration or aerosol extinction is constant below the layer height. However, this is not the case if the shape parameter is larger than 1. Please correct this.

Author reply: We added in brackets: (except for S> 1, see below)

p.3904, line 24: "of " should be "or".

Author reply: corrected

p.3905, line 1-5: I do not understand the sentences "Exponential profiles ..."

Author reply: Probably the misunderstanding resulted from the phrase 'with only one parameter'. We removed this phrase and hope the statement is more clear now.

p.3905, line 12-13: Why do both possibilities fail if smooth vertical gradients have to be described?

Author reply: Because of the discontinuity between the two layers. We added this information to the text.

p.3907, Eqs. (8) and (9): It seems to me that the equations should be "AOD/L(2-S) or VCD/L(2-S)". Please check them.

Author reply: Many thanks for this hint! We corrected both equations.

p.3912, line 17: Please revise the sentence "Fortunately ..." to be more quantitative one.

Author reply: We changed the text to Fortunately, for most measurements the retrieved AODs only slightly depend on the assumed shape parameter: taking into account all observations, for 74% the difference in the AOD is below 20%. If only clear observations with layer height (S=1) < 1.2 km are considered, for 97% the difference in the AOD is below 10% (see also Fig. 4).

p.3914, line 23: It is unclear to me what the results of the radiative transfer simulations presented in Fig.5 indicate here.

Author reply: We changed the text to: 'There is probably no simple explanation for this finding, but the fact that L1.1 is systematically smaller than L1.0 is also supported by the results of Fig. 6 (and Fig. S4 in the supplement), where the O4 DSCDs for profiles with S = 1 and L = 1 km agree with those for profiles with S > 1 and L > 1 km'

p.3915, line 4: I also think that aerosol retrievals with S = 1.1 is not a good choice, because it requires a correction factor as high as 2.

Author reply: We changed 'good (pragmatic) choice' to acceptable (pragmatic) choice'

p.3915, line 8-10: It is stated here that the results of the trace gas inversions are more realistic and consistent, if S<1 for the aerosol profile inversion is chosen. However, on p.3914, line 14-17, the authors mention that stable results for AOD have not been obtained for all days with S<1. I think that using different S(aer) values for aerosol and trace gas inversions to achieve good agreement for both cases would be unfair.

Author reply: The apparent contradiction can be well justified. As described in the paper, the choice of S>1.1 is an acceptable pragmatic solution for the retrieval of the AOD. For values of S < 1 the AOD would be often overestimated. However, if the aerosol extinction profiles for S = 1.1 were also used as input for the trace gas profile inversion, a particular problem occurs: the aerosol extinction close to the surface would be systematically underestimated in most cases, while the maximum trace gas

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concentrations are typically located at these altitudes. To avoid this problem, we use aerosol extinction profiles retrieved for a shape parameter S £ 1. Even if in some cases the AOD (and the aerosol layer height) would be overestimated, the aerosol extinction close to the surface will very probably be more correct than that for aerosol retrievals with S = 1.1. We added this information to the text.

p.3915, line 20: It would be better to say that "... the dAMF(alpha) values for NO2 and HCHO do not depend on the absolute value of VCD, ...".

Author reply: Many thanks for this suggestion. We changed the text accordingly.

p.3915, line 27: Why the absolute value of dAMF can be obtained from the comparison between normalized (i.e., relative) quantities dSCD and dAMF?

Author reply: Because of the unique relationship between normalised dAMF and the absolute dAMF, from which the normalised dAMF were calculated. We added this information to the text.

p.3918, line 15-17: For the light path along the instrument line of sight to increase, the cloud bottom should be higher than the altitudes, where the last scattering by air molecules occurs under clear sky conditions. This condition should be mentioned here.

Author reply: We added this condition to the text.

p.3919, line 7: "... the dAMF(alpha) are ..." should be "... the dAMF(alpha) values are ..."

Author reply: corrected.

p.3919, line 12-25: The profile inversion is used to investigate the diffusing screen effect. However, I am skeptical that the used profile inversion is suitable for this purpose, because it may be unstable. Was the layer height L stable for all cases investigated here? I expect that aerosol inversion tends to retrieve AOD that contains a contribution from cloud optical depth assumed and may show overestimation. Why doesn't this

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tendency appear in Fig. 11?

Author reply: As stated before (and also stated in the revised version of the manuscript, in section 3.4), the aerosol inversion shows only negligible dependence on the initial values. This holds for both observations under both clear and cloudy skies, and also for the results of our sensitivity studies. The 'contribution' of the cloud OD to the retrieved AOD depends on the height of the cloud bottom. For cloud bottom heights > 2km (lowest cloud bottom height tested), clouds tend to underestimate the AOD. This is caused by of the diffusing screen effect described in detail in section 4.1.1. As suggested, we added the retrieved layer height to Fig. 12 (Fig. 11 of the original version). Like the AOD, they also depend on cloud height and cloud OD.

p.3921, line 1: I understand that the colour index is less sensitive, but does it give additional information?

Author reply: In principle the CI could give additional information, because the effects of aerosols and clouds on the CI are different. However, the detailed interpretation of the CI would require more comprehensive radiative transfer simulations. We added this information to the text.

p.3925, line 18: What are the parameterised MAX-DOAS errors?

Author reply: The parameterisation of the MAX-DOAS errors was described in section 3.3 and table 3. We added this information to the text.

p.3927, line 13: Perhaps a better word is "available" than "possible".

Author reply: corrected

p.3928, line 21: Please delete "a" just before "rather".

Author reply: corrected

p.3931, line 10: It seems to me that the word "true" is too strong to be used as a value obtained by inversion.

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Author reply: We replaced 'true ' VCD by 'VCDs obtained by the profile inversion'

p.3931, line 16: It would be better to explain "systematic dependence" quantitatively, for example, using the correlation coefficient, slope, and intercept.

Author reply: The details of the correlation analyses are given in Fig. S11 in the supplement. We added this information to the text: '. . .(for details of the correlation analyses see Fig. S11 in the supplement) '

p.3934, line 26-29 and p. 3935, line 1, 9-12: What do the ranges of agreement (+/-) represent?

Author reply: We estimated the consistency between the different telescopes from the slopes of the correlation analysis and the ratios between the respective results (see section 5.2). We added this information to the text.

p.3935, line 9: Would it be better to mention the agreement for aerosol extinction as well?

Author reply: We added a comparison of the average aerosol extinction to Fig. S7 in the supplement (Fig. S4 in the original version). We also included the comparison results at the end of section 5.2.1. (Note that also Table 3 was updated and includes now error estimates for the retrieval of aerosol extinction).

Interactive comment on Atmos. Meas. Tech. Discuss., 4, 3891, 2011.