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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.

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General comments: This article describes aerosol and trace gas (NO2 and HCHO) results from MAX-DOAS measurements over Milano, Italy in September 2003. Spatial heterogeneity of the aerosol and gas pollution was studied by simultaneous MAX-DOAS measurements in west, north and south directions. The authors present a parameterized inversion algorithm to retrieve trace gas total vertical column (NO2 and HCHO)/aerosol optical depth, layer height, and profile shape parameter. Sensitivity studies were conducted to evaluate effect of profile shape parameter on the retried total gas column/AOD and layer height. A cloud classification and screening algorithm is introduced to identify cloudy and clear sky conditions. Effect of clouds on the retrievals was studied. Extensive correlation analysis was conducted to compare MAX-DOAS results with other independent measurements. This paper addresses a very important question of the profile retrieval from MAX-DOAS measurements. While optimal estimation is widely used in such retrievals, it requires a priori knowledge about the profile distribution and its variability. In most cases this information is not available, and a priori profile is assumed. Optimal estimation often retrieves unrealistic (negative) concentrations. As with any other method the solution is not unique (due to limited information in MAX-DOAS measurements). Attempt to use parameterization of the profile is an alternative solution that does not depend on the a priori information (although initial parameter guess plays an important role) and does not produce negative concentrations. Understanding limitations and possibilities of different parameterization scenarios is very important. I believe this paper fits the goals of AMT and recommend publishing the paper after some revisions. In general, the paper is well written and organized. The main confusion, however, arises in sections 3.4 and 3.5. Specific comments and technical corrections are listed below.

Author reply: We thank the reviewer very much for the positive assessment and the recommendations for improvements. Before we respond to the suggestions point by point, we want to briefly introduce the main changes of the revised version of the manuscript.

-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 unsta-



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ble 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). 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.

Our detailed answers to the specific points are listed below.

Specific comments and technical corrections:

3900, 15 -18. DSCD retrieved relative to a single (fixed) reference spectrum also will reflect changing photon path due to solar movement (solar zenith angle).

Author reply: Many thanks for this hint. We now also mention the effects of a changing solar zenith angle.

3900, 29. Please specify aerosol loading from AERONET at 340 nm



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Author reply: The AOD at the AERONET station at Ispra was 0.14. We added this information to the text.

3901, 17 – 18. Please clarify if the O4 correction factor of 0.75 was derived from the data collected during this study or taken from Clemer et al. 2010

Author reply: The correction factor was derived from our own measurements (see Wagner et al., 2009). We changed the text to 'This correction was found to be necessary to bring OUR model results and measurements under almost aerosol and cloud free conditions into agreement (see Wagner et al., 2009, Clémer et al., 2010).' We also saw that we mentioned a wrong correction factor in the original version: instead of 0.75 we use 0.79. We changed the text accordingly.

3901, 26 – 27. Please rephrase the second part of the sentence.

Author reply: We changed the text to: 'For the interpretation of the profiles retrieved from the MAX-DOAS observations it is important to know the spatial distribution of the sensitivity of the MAX-DOAS observations. ' 3902, 1. Please clarify: measurement sensitivity to what?

Author reply: We meant the measurement sensitivity to aerosols and trace gases. We added this to the text.

3902, 3 – 6. Please explain how you estimate 5 km distance. Do you expect homogeneity condition to hold along this distance at the measurement site?

Author reply: We added new radiative transfer simulation results to the manuscript (and a new Figure S1 in the supplement). They are made for different assumed aerosol and trace gas layer heights (and for different SZA and relative azimuth angles). For these simulations we assume horizontal homogenous layers. The horizontal sensitivity ranges are between 1.5km and 22km.

We changed the text in the main part of the manuscript to: For the interpretation of the profiles retrieved from the MAX-DOAS observations it is important to know the horizontal range, for which the MAX-DOAS observations are sensitive: the larger the sensitivity range is the higher is the probability that horizontal gradients affect the profile inversion. The measurement sensitivity to aerosols and trace gases depends on the distance from the instrument location, varies with several parameters (e.g. viewing geometry, wavelength, aerosol and trace gas profiles), and is thus difficult to quantify. Also there is a systematic geometric relationship between the probed altitude and distance for each elevation angle: The sensitivity for the lowest atmospheric levels is highest close to the instrument. In the supplement the horizontal range for which the MAXDOAS observations are sensitive sensitivity are estimated for various conditions. I typically ranges between a few kilometres and about 20 km. Note that in our inversion algorithm horizontal homogenous conditions are assumed.

We put a description of our method to estimate the horizontal range for which the MAX-DOAS observations are sensitive to the supplement. We also added the respective results (Fig. S1).

3904, 24. Replace: Either convex “or” concave.

Author reply: corrected

3905, 5. Please explain how dSCD measurements at multiple wavelengths can provide additional pieces of information about the profile shape.

Author reply: Especially close to the surface the sensitivity of MAXDOAS measurements for low elevation angles depends strongly on wavelength. These differences can be exploited to improve the accuracy of the profile inversions. Detailed studies about the information content were performed by Frieß et al., 2006. We added this reference here.

3905, 6. “Two layer profiles” term is a little misleading since only one elevated layer is retrieved.

Author reply: We added the following ’warning’ to the text: ’Note that the term ’two layer



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profile' is not fully appropriate for the chosen parameterisation with only the amount of one layer freely fitted (and the amount of the other fixed to zero). However, we keep this term throughout the manuscript in order to be consistent with future measurements (with a higher information content), from which amounts of two layers could be independently determined.'

3905, 15. Please clarify value of which parameter is fixed: "For the two-layer profiles we fixed the value of the lowest layer"

Author reply: Probably the reviewer has overlooked that already in the original manuscript the value (zero) was given in the text.

3906, 16. What is the assumed shape parameter (S) under "unstable" inversion conditions?

Author reply: Since the fixed values of the shape parameter S is discussed in detail in sections 3.4 and 3.5, we added a hint to these sections to the text: 'In such cases one of the profile parameters introduced above (the shape parameter, S) is set to a fixed value (for details see sections 3.4 and 3.5).'

3907, 24 – 25. According to eq. 4, Eq. 8 and 9 should be: Ave AOD (or Ave VCD) = AOD (or VCD) / [L*(2-S)]

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

3908, 5. Please specify the wavelength at which modeling simulations are performed

Author reply: To minimize the computational effort, all simulations were performed at 360nm. This wavelength is well suited for the interpretation of the O4 absorption at 360nm. For the NO2 and HCHO observations, simulations at a slightly smaller wavelength might have been more appropriate. We estimated the errors due to the shift in wavelength by comparing selected simulation results for 350nm with those for 360nm. The differences are rather small (typically below 3% and for AOD >0.3 below 1%). We added this information to the text.



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3911, 17. In the first step of the “trace gas” profile inversion

Author reply: corrected

3912, 7. Is your statement “that the information content of our MAX-DOAS observation is not sufficient to discriminate these different profile shapes” applicable to all your measurements during the campaign or just to 15 and 19 September, 2003? It is expected that different AOD profiles might produce similar O4 dAMF. Have you made an attempt to identify the “common” conditions under which this happens?

Author reply: Such ambiguities can be caused by different reasons, e.g. by elevated layers, horizontal gradients or the influence of clouds. For about 60% of all MAX-DOAS measurements during the FORMAT-II campaign stable profile inversions of all three profile parameters were possible: The inversion results did not significantly depend on the initial values and that they did not rapidly change between succeeding observations. Most of the unstable inversions were found for observations under cloudy skies. Interestingly, even for the example in Fig. 3 for 19 September 2003 the fit found a meaningful inversion result (for a shape parameter of 0.97), although the RSS value for this solution is only slightly smaller than for other shape parameters. We added this information to the text.

3912, 11. You probably can make a general statement here that behavior on 15 Sep is representative of elevated aerosol layers (lower ext. coefficients at the surface than aloft) and 19 Sep is typical for aerosols located mainly close to the surface (based on the sensitivity studies).

Author reply: Many thanks for this suggestion! We added the following information to the text: The O4 dAMFa on 15 September 2003 indicate the presence of elevated aerosol layers (see below), while the O4 dAMFa on 19 September 2003 are representative for aerosol profiles with maximum extinction at the surface. Here it should, however, be noted that the details of these dependencies also vary with SZA and relative azimuth angle.



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3912, 17. How do you choose the "correct" profile from your inversion? While on 19 Sep 2003 the AOD agree (within 20%) for different profiles the actual fit of the measurements to the forward model is not as good as than on 15 Sep 2003. What are the "typical" differences between the AODs retrieved using different shapes for clear, similar atmospheric conditions?

Author reply: For many observations it is difficult to chose the correct profile shape from the inversion results. We investigated the typical differences for AOD retrievals using different shape factors. For clear sky conditions and for layer heights below 1.2 km (1.0km) compact linear relationships between the results for shape parameters S = 1.0 (S = 0.8) and those for S 1.1 were found with slope of 1.17 (1.22) and rˆ2 of 0.99 (0.98). Larger deviations occurred for retrievals with higher layer heights. We added this information and a new figure (Fig. 4) to the manuscript.

3912, 22. Please replace "observations" with "retrieved parameters"

Author reply: We agree that 'observations' is not a good term here. But we replaced it by 'profile retrievals', which we think is a slightly better choice than 'retrieved parameters'.

3913, 23. Could you please explain your choice of SZA 30_ and RAA 0_ for your hypothesis testing? Modeling results using RAA of 0_ and small relative zenith angles (in this case e.g. 12_ for 18_ elevation angle) might be difficult to test with MAX-DOAS observations. External stray (unwanted) light entrance from Sun into a MAX-DOAS instrument is possible at such a small RAA and RZAs. Since these "stray" photons travel a shorter path than the "properly" scattered photons the resulting O4 dAMF is lower. Such data will result in higher AOD retrievals. In addition, aerosol forward scattering is very sensitive to aerosol phase function.

Author reply: We agree that simulations for viewing directions close to the direction of the sun are probably not fully representative for observations at different directions. However, for the chosen geometry these problems should still be small, because the



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smallest angle between the viewing direction and the sun is still larger than 30°. Nevertheless, we performed additional simulations for other viewing geometries, which supported the results shown in Fig. 5 (Fig. 6 in the revised version of the manuscript). (see also reply to the next comment)

3914, 1. I believe you need more forward model simulations for other aerosol types and profile parameters, and relative viewing geometries to support your conclusion here. Please also provide your "confidence" in the modeling results for 1deg viewing elevation angle.

Author reply: We performed additional simulations for other viewing geometries, profile parameters and aerosol optical parameters. The simulated O4 DAMF were then used as input of our profile inversion (using shape parameter S = 1.0). The retrieved AODs and layer heights were found to be systematically larger than those used in the forward model confirming that the results shown in Fig. 5 (new Fig. 6) are of general relevance. We have no reason to doubt our simulation results for 1° elevation, since we use a full spherical Monte Carlo model.

On Fig. 5 please add shape parameters (S = 1) for red and green curves.

Author reply: Corrected

3914, 14. From your discussion here, I conclude, that the shape parameter S = 1.1 provides the most stable AOD retrieval but you believe that S = 1 is physically more realistic, so you perform the S=1.1 retrieval and then make it look like S = 1 by adjusting the L. If this is correct please rephrase.

Author reply: We added the following sentence: For many days (without elevated profiles) we use an assumption which is obviously wrong. As a consequence, the retrieved AOD is often smaller than for shape parameters S £ 1 (see Fig. 5 right), but fortunately this underestimation is usually small: For 74% of all observations it is less than 20%; for 97% of clear observations with layer height (S=1) < 1.2 km the difference in the



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AOD is below 10% (see also Fig. 4).

3914, 17. Not sure why you are referring to Fig. 4 here.

Author reply: We changed the hint in the brackets to '(results for one day are shown in Fig. 5 left)'

3915, 8. This sentence is somewhat confusing. It gives an impression that O4 analysis of aerosol profiles using your parameterization technique with the pragmatic approach is not useful for the trace gas analysis. Since the final goal is trace gas profile inversion it is not clear why you discuss the pragmatic approach at all.

Author reply: We changed the text at the end of section 3.4 to: The different choice of the shape parameter S for either the retrieval of AOD or the retrieval of trace gas profiles might be seen as an inconsistency. However, we think these choices are well justified. As discussed above, the using a shape parameter $S > 1.1$ leads to more consistent results of the AOD than the use of S £ 1. 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 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.

3917, 20. If the average VMR is independent of the profile parameter, and assuming all of the NO2 is in PBL, would the information about the PBL height help determining which profile parameter to use to obtain the "correct" VCD?

Author reply: Maybe we misunderstand this suggestion, but we think it contains an inconsistency: For the calculation of the average VMR not only the partial VCD below



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the PBL (layer height L) is used, but the total tropospheric VCD including the exponential decrease above L. For S < 1, this part contributes to the total tropospheric VCD. If only the partial VCD below the PBL was used for the calculation of the average VMR, it would not be independent of the assumed shape parameter. This holds only if the total tropospheric VCD is used.

3919, 11 What is the cloud height and thickness?

Author reply: For the simulations shown in Fig. 10 the cloud height is between 4 and 5km and the cloud OD ranges between 1 and 5. We added this information to the text.

3921, 3. Can you be more specific? Which wavelengths would you recommend?

Author reply: Many DOAS instruments cover the full visible spectral range. The colour index calculated from such observations will provide a stronger contrast. We added the recommendation to use the whole visible range to the text.

3923, 17. Could you summarize which aerosol and trace gas profiles (shape parameters) were used in retrieving the final results?

Author reply: We added the following information directly before section 5.1. Note that all AOD results shown in this section were obtained using a shape parameter of 1.1 (see section 3.4). All trace gas results were obtained using a shape parameter of 1.0 both for the aerosol and trace profile inversion.

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