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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Multi Axis Differential Optical Absorption Spectroscopy (MAX-DOAS)" *by* G. Hönninger et al.

G. Hönninger et al.

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Response to referee comment RC S2237 (anonymous referee 1) from December 18, 2003:

We very much appreciate the detailed and helpful comments by the referee, which helped us improving the clarity of this paper.

General comment 1: Anonymous referee 1 suggested that we add more information about the instrumentation.

Response: We agree with the referee and expanded our instrumental descriptions, see below for details.

General comment 2: The referee also raises a question about uniqueness of the solution of the profile retrieval algorithm asking whether the solution obtained from the

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algorithm might just be one of several solutions that cannot be discriminated against.

Response: This comment is very much related to the general comment of A. Maurellis (see also Response to A. Maurellis below). In fact our current approach to retrieve profile information from our MAX-DOAS measurements is not yet a fully developed inversion algorithm. What we rather described in the paper is a "conceptional algorithm". In a forward model we calculated the dependence of the apparent slant column densities on the elevation angle for various profiles. We also performed sensitivity studies for various parameters of the radiative transport in order to determine the quality of the results. This approach essentially describes a "grid search" method, where the "best fit" profile is chosen among a set of profiles calculated by varying all

possible parameters. The choice of the "best fit" forward calculated profile as the actual profile is described for example in Section 6.1 and Figure 17 and certainly allows discriminating the "best fit" against the other forward modelled profiles. The possibility that another profile, which is significantly different from the "best fit" also agrees with the MAX-DOAS measurements appears to be extremely small. The future application of a full inversion algorithm will eventually allow to express the errors involved in the determination of the actual profile in a more detailed way.

Specific comments: As suggested by the referee we included the phrase "in the atmosphere" in the first sentence of the abstract. The referee also asks what is meant by "close to the instrument". Using the phrase "close to the instrument" we intended to express both, that a ground-based instrument is most sensitive to absorbers near the ground and an airborne instrument would be more sensitive to absorbers in the vicinity of the platform's cruising altitude. In the original paper we then gave a more precise sensitivity range for ground-based MAX-DOAS with the phrase "the lowest few kilometres of the atmosphere". Moreover, at the end of the introduction we explain "close to the instrument" as "in the boundary layer with ground-based instruments". We agree that this statement might still not be very precise, however, actual numbers are given in the respective sections, good accuracy is obtained in the lowest 2km (profiles P1-P4,

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sections 4.1-4.7), whereas the sensitivity decreases in the free troposphere for profile P7 and P8 as discussed in Section 4.8.

As suggested by the referee we include the references to Wahner et al., JGR94, 16619, 1989 in table 1, Harder et al. 1997 on page 5597 and Kattawar et al. 1981 on page 5602 in the revised manuscript.

As suggested we also added an explanation on page 5597: "This is due to the fact that the differential absorption pattern of the trace gas cross section is unique for each absorber and its amplitude can be readily determined by a fitting procedure using for example least squares methods to separate the contributions of the individual absorbers."

We changed "average" to "path average" as suggested on page 5603:

In the radiative transfer calculation section on page 5612 we clarified our statement and in order to include the possibility of inhomogeneous distributions we changed the beginning of the paragraph to: "In the case of horizontally inhomogeneous trace gas distributions the observation azimuth angle is clearly of importance and has to be determined in each individual case. For horizontally homogeneous distributions a dependence of the air mass factors on the relative azimuth..."

Regarding our Section on "Practical realisation" the referee suggested moving Section 5.1 to the section describing the DOAS technique. We think that our original order of the manuscript is preferable as we first explain the DOAS technique, then the theory of MAX-DOAS and subsequently come to the section explaining the practical realizations. We agree, however, that this section needed some improvement which we addressed in the following way: We added more technical details and descriptions on the individual systems and include a new table summarising all the necessary specifications of the various set-ups. We also explain in more detail where the problems with certain instrumental set-ups originate and how they can or why they cannot be resolved. We added more details to the description of scanning and simultaneous MAX-DOAS as suggested by the referee. We particularly describe how "cross-convolution" - al-

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though it can be readily implemented and calculations can be done quickly - affects the sensitivity of the measurements: "none of these approaches has yet provided the same sensitivity as can be reached with a scanning instrument. Any convolution of measured atmospheric spectra with an instrument function of typically 5 - 10 detector pixels FWHM reduces the spectral resolution and essentially "smoothes" intensity variations (e.g. spectral features, photon shot noise, detector pixel-to-pixel variability and electronic noise) that are below the spectral resolution (FWHM) of the instrument. This can result in spectral artefacts due to smoothing of noise and thus can affect the quality of the DOAS fit (e.g. Bossmeyer, 2002). While detector pixel-to-pixel variability can largely be removed by correcting with a "white light" spectrum (e.g. from an incandescent lamp), taking these additional spectra regularly comes at a cost of measurement time additional power requirements and experimental effort."

In the revised version we also discuss in more detail the advantage of simultaneity/ disadvantage of non-simultaneity in the description of the scanning instrument: "It certainly depends on the respective aim of a field study whether this represents a significant drawback. For example, at high latitudes simultaneity may often not be an issue because the SZA changes only slowly there. In contrast to that the effect of changing SZA and thus the observed stratospheric trace gas column densities is much more pronounced at mid- and low latitudes. However, since the change in SZA can be well described by a simple polynomial fit to the measured time series for each elevation angle, its effect can be removed by interpolating all time series on a common grid prior to further analysis (Leser et al., 2003). On the other hand interpolation can be difficult during periods of rapidly varying radiative transport conditions in the atmosphere, for example varying cloud cover, aerosol load, during sunrise/sunset or on a moving (e.g. airborne) platform. In the latter cases and when high time resolution is required simultaneous observation should be preferred (see below)."

Referee 1 noted that error bars in Fig. 21 would help. Indeed, error bars are already present in this figure. While the quality of printouts will certainly vary from printer to

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printer, the best way of looking at this figure is actually in the electronic (pdf) version, where the error bars are clearly visible. The time resolution for the two measurement techniques (MAX-DOAS: 5 min for one single measurement, 30 min for a complete MAX-DOAS scan, with somewhat longer times during morning/evening twilight periods; LP-DOAS: 30 min-1 h) is stated in the text in the revised version and can also be seen in Figure 21.

The referee suggested a more detailed discussion of nonhomogeneous air plumes and asks how the effect of non-homogeneous plumes is removed and if results in the non-homogeneous case can be believed.

MAX-DOAS as a technique that averages over an absorption path of typically hundreds of metres up to more than 10km, cannot resolve inhomogeneities in concentration that are present along the lightpath. The combination of several viewing directions allows deriving vertical profile information to a certain extent, however, it cannot be expected that MAX-DOAS can resolve strong inhomogeneities that occur on much smaller scales than the typical resolution of MAX-DOAS of 500m-1km. On the other hand spatially averaged data may be much more suitable to characterise pollution levels than highly variable data from an in situ instrument in case of inhomogeneities. We addressed this by including statements on the effects of inhomogeneity in the appropriate sections of the paper: Section 8.1 "Possible local inhomogeneities including vertical profiles can be studied by MAX-DOAS observations using multiple viewing directions, for example observing light at different azimuth and elevation angles."

Section 8.2 "In case of inhomogeneous trace gas distributions frequently found near emission sources in urban regions retrieving profile information from MAX-DOAS measurements is further complicated. However, average trace gas levels derived from MAX-DOAS measurements are less likely to be affected by non-homogeneous distributions than localized measurements using in situ instruments. A further advance on scattered sunlight DOAS, the so-called "Imaging DOAS" (Lohberger et al. 2004), which can be applied to study for example non-homogeneous plumes by taking two

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dimensional images of trace gas distributions."

End of Section 8.2 "Compared with localized measurements at the crater rim or airborne measurements within the plume, MAX-DOAS is not sensitive to inhomogeneities of the plume but rather averages across the absorption path. Temporal variability of emission patterns can be studied by automated MAX-DOAS measurements over a longer time period."

References The web site given for the Hönninger thesis is correct and accessible. However, at closer look a blank character in the PDF-file produced for ACPD appears to prevent the common method of copy/paste to a web browser from working in this case. The link: http://www.ub.uni-heidelberg.de/archiv/1940 should be typed without any blank character. This typo will hopefully not occur in the final version.

Response to referee comment RC S1945 by A. Maurellis from November 18, 2003

1 General Comments: We very much appreciate the comments by A. Maurellis which helped us improve the clarity and quality of our paper. This reviewer's general comment that we barely made the connection to profile retrieval techniques used in airborne and space-borne remote sensing is correct. We addressed this by including references to these techniques in the updated version of the paper. See also our response to the specific comments below.

2 Specific Comments

A. Maurellis pointed out that several the spectroscopic details of the various instruments used for our measurements were not mentioned in the paper. This is true, and we realize that readers will appreciate to see a list of these details which one had to look up in the referenced literature otherwise. We addressed this by including a more comprehensive description of the spectroscopic properties of the instruments employed, which are summarized in an additional table (Table 4, see also response to anonymous referee 1). We also include a table summarising the details of the simulations

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we performed in this paper (Table 3). Regarding the referee's wish to see a spectral fit or even a range of spectra as a function of elevation angle we intended to focus on the MAX-DOAS technique in this paper rather than repeating basic DOAS column retrieval from spectral fits. The best illustration on how the measurement signal changes with elevation angle can probably be seen in the Bobrowski et al. 2003 paper, where the absorption signals of SO2 and BrO clearly increase towards the centre of the plume, reaching background levels away from the plume. We feel that it is not necessary to repeat this quite obvious finding, that increasing DSCDs correspond to increasing absorption signal/fit coefficient in the spectral fit.

A. Maurellis was also wondering about the way we did our mathematical inversion. This is unfortunately the result of a misunderstanding due to the misleading way section 4.5 was written in our paper. We agree that from the way it was written the reader could assume that the actual inversion algorithm is already fully implemented and working. This is not the case and therefore we re-wrote section 4.5 in order to avoid this misunderstanding, although it may now sound a bit awkward (which was the reason why we used indicative forms in the first version). However, it now properly accounts for the fact that we intend to implement this algorithm in future applications. At present the algorithm only works by manually choosing the "best fit" from a limited number of forward modeled profiles calculated for a series of parameter values.

The misunderstanding explained above largely prevents us from answering the following questions brought up by A. Maurellis. We can only answer them thoroughly once the inversion algorithm has been fully implemented and the results interpreted. For now, we have to admit that the way we derive profile information is solely based on a "grid search" method where we compare the measurement results with a set of simulated forward modeled profiles and choose the best fit as for example described in Section 6.1 and Figure 17. In order to make this clear to the reader we included the following statements in the revised version of our paper: At the end of the introduction: "While the approaches developed in inversion theory (e.g. Rodgers, 1976) can

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- and will in the future - be applied to MAX-DOAS, no mathematical inversion was attempted in this study. Profile information was rather derived from the comparison of measurements and various forward modelled profiles." Beginning of RTM section: "An actual profile inversion algorithm (e.g. Rodgers, 1976) was not yet implemented for MAX-DOAS at the current stage. Instead we apply the approach to perform a series of simulations for a number of possible profiles and subsequently choose the "best fit" as the most likely profile."

3 Technical comments Section 7, second paragraph We corrected the mistake and now refer in all cases to figure 21.

Figure 18 actually displays error bars in grey color. We admit that these are not self-explanatory unless the pdf-file of the figure is viewed at large magnification. We therefore added to the caption the following statement: "Error bars (2σ) are shown in light grey."

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