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Interactive comment on "MAX-DOAS measurements of formaldehyde in the Po-Valley" *by* A. Heckel et al.

A. Heckel et al.

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We appriciate the helpful constructive comments of referee #2. These comments helped us improving our paper. Most of the more technical comments were incorporated in the final version as suggested. In addition there were some more general comments which we want to discuss here:

RC: General comments However, in my mind the manuscript also raises a couple of questions: First, if the technique is so strongly depended on cloud-free days, how useful is the whole data set and how many days (e.g. out of the 30 days of measurements) can in reality be used for the data analysis as presented in this paper - maybe not many more than the 3 days discussed in this publication?

AC As for many atmospheric measurement techniques, clouds are a problem for MAX-DOAS measurements, in particular if profiles are to be retrieved. In principle, clouds can be simulated in the radiative transfer, and by comparison with the O_4 measurements, the results can be iterated until a good representation of the cloud is reached. In practice however, clouds are often complex and change rapidly over time, and even a good simulation will introduce large uncertainties in the derived profiles.

RC Second, to retrieve more reliable profile values and, in turn, boundary layer mixing ratios, a proper inversion technique such as optimal estimation is most likely necessary for the proper interpretation of the multi-axis measurements. This addition would clearly improve the data analysis presented in this study and allow a more quantitative interpretation of the results.

AC This publication was thought to be a first approach to show possibilities and value of these kind of measurements. In this study, the optimal trace gas profile was determined by a simple selection process where many different profiles were used to compute vertical columns for the different viewing directions and the profile that leads to the best agreement between the different viewing directions was chosen. This manual approach has of course to be replaced by a more systematic evaluation based on an optimal estimation algorithm.

RC Third, how realistic is the assumption of horizontal homogeneity in this study and if this assumption doesn't hold, what potential impact has that on the results (VCDs, profiles and boundary layer mixing ratios)? The authors themselves raise the issue and point out that horizontal inhomogeneities can have a large impact and lead to incorrect vertical profile retrievals. How sure can we then be, that the presented results are reasonable given that the assumption of a horizontally homogeneous distribution of HCHO over 40 km has to be made when the sources are only about 50 km from the measurement location?

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AC There is different impact on the results. While the retrieved vertical columns are only affected within the range of the error given, there is quite big impact on the retrival of profiles and mixing ratios by horizontally inhomogeneity. But as seen from figure 7 these effects can be seen just by looking at the temporal evolution of the vertical columns of the viewing directions. Inhomogeneities which are constant with time can possibly not so easily be seen and lead to potential errors. With respect to the numbers given in the text we have to admit, that the value of 40 km given for the order of homogeneity was an unfortunate mistake. It just respresents the geometrical light path in an 2km boundary layer with an view angle of 3°. An estimated length of the total light path would be in the order of 15 to 20 km, according to AMF calculations. But this is the total light path in the boundary layer. Due to multiple rayleigh scattering in the UV this is not an estimte for the distance the light is coming into the instrument.

RC: Page 4, column 2, line 1-2: How can you be certain that there were no clouds during these 3 days (maybe there were hardly visible clouds) that could have disturb the analysis?

AC: We can see no indication of cloud influence in the data (i.e. the individual diurnal time series of intensity and O_4 absorption signal usually show cloud influence by strong temporal variations). In addition we think, that if there are clouds which affect the retrieval, it would be noticeable during the adjustment of the aerosol parameters, leaving larger deviations between the signals from the different elevation angles.

RC: Page 6, column 1, line 31: 500 m seems to me to be very high resolution given the technique. And since no optimal estimation technique was used, how can you be sure that your resolution was around 500 m and e.g. not rather 3 or 5 km?

AC: The vertical resolution of the profiles strongly depends on altitude. As shown in Wittrock et al. (2004), the viewing directions close to the horizon have a strongly

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peaking sensitivity for the lowest 500 m, and although no quantitative evaluation of the averaging kernels has yet been performed, simulations show that profile changes in the lowest 500m and in the lowest 1000 m have significantly different effects on the columns observed in the lowest viewing directions. Above this lowest layer, the vertical resolution is much reduced, and we estimate it to be of the order of 3 and then 5 km.

RC: Figure 9: What causes the discontinuities in the MAX-DOAS data set when the actual resolution of the measurements is around 5 minutes? Is it cloud conditions? Also, you could indicate representative error bars for a couple of data points for the different data sets.

AC: In order to explain the discontinuities we found, that the sentences at page 6, column 1, lines 37-39 we phrased misleading and unclear. We change these two sentences to: "For each of the highlighted areas in figure 7 one profile was retrieved, assuming no major changes in the aerosol and in the shape of the trace gas profile during these time periods. The VMR values within these periods result of scaling the a priori trace gas profile to match the measured vertical column. The discontinuities seen in figure 9 show the limits of this approach to use one set of AMFs valid for one data point for a larger time period."

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 1151, 2004.