Atmos. Chem. Phys. Discuss., 6, S6584–S6587, 2007
www.atmos-chem-phys-discuss.net/6/S6584/2007/

Atmospheric Chemistry and Physics Discussions

ACPD

6, S6584–S6587, 2007

Interactive Comment

Interactive comment on "Spatially resolved measurements of nitrogen dioxide in an urban environment using concurrent multi-axis differential optical absorption spectroscopy" *by* R. J. Leigh et al.

Anonymous Referee #2

Received and published: 30 January 2007

The manuscript by Leigh et al describes measurements by a multi-axis Differential Optical Absorption Instrument in the urban environment of Leicester, UK. The observed trace gas slant column densities are converted to concentrations based on a number of simplifying assumptions. The observations of NO_2 are validated by comparison with measurements from air quality monitoring stations in the city. The temporally highly resolved slant column densities, together with a simple description of the light path geometry, were also used to investigate the spatial extent and NO_2 concentration of plumes. The authors conclude that their instrument is able to quantify

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Interactive Discussion

Discussion Paper

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the concentration of NO_2 in an urban area and the NO_2 content of a single NO_2 plume.

This paper presents an interesting data set of MAX-DOAS observations in an urban area. The interpretation of the temporal multi-axis data is an interesting idea which deserves to be explored further. However, the paper lacks thoroughness, and major issues with the data interpretation need to be addressed before publication in ACP.

First and foremost, no uncertainties or errors of any of the presented data are given. It is therefore impossible to assess the author's statements on the accuracy of their instrument. One of the main conclusions of the manuscript is that the instrument provides useful measurements of spatially integrated concentrations (page 12686, lines 17 - 22). Without an analysis of the uncertainty of the calculated concentrations, this statement cannot be made.

The conversion of slant column densities to concentrations is based on extremely simplifying assumptions that are insufficiently justified. Assumption 1 states that the trace gas concentrations above the planetary boundary layer (PBL) are the same for zenith and off-axis scan. However, the authors use a noon-time zenith reference for their analysis. It is hard to conceive that the NO_2 concentration and slant column density above the PBL did not change throughout the day. It should be noted that typically MAX-DOAS application use a temporally close zenith scan to overcome this problem. The authors should explain what the advantages of a noon-time zenith reference is, or use a temporally close zenith spectrum to avoid the uncertainties introduced by temporal changes. Assumption 2 states that clouds are assumed to be present as a uniform layer above the PBL. This is obviously not true and deserves more justification than given in the manuscript. The choice of a 2 km absorption path in assumption 3 and 4 is arbitrary. Geometrically this would mean that for a 5° elevation angle absorption solely occurs in the lowest 200m. Hönninger et al. (2004), give box airmass factors of 10 for the lowest 500m of the atmosphere based on radiative transfer calculations.. Using these box-airmass factors the absorption path in a 500m high PBL would be around 5km. One can construct a case where urban haze will decrease the effective pathlength for a higher PBL, but it is hard to imagine that the aerosol levels and the PBL height in Leicester do not change on a day to day basis. The authors have to provide an uncertainty analysis for this assumption. How much does the calculated concentration change upon changes in aerosol extinction and PBL height?

To make the comparison of ground-based air quality monitoring observations with the MAX-DOAS measurements meaningful the uncertainty by the above assumptions need to be given. Alternatively the authors could use a radiative transfer model and meteorological data on PBL height to provide a more quantitative analysis of the observations. The authors have the expertise to do such an analysis as shown by a recent publication (Frieß et al., 2006).

The analysis of the temporal behavior of the observed slant column densities is the most interesting part of the manuscript. Extracting data on the size, origin, and the total NO_2 content of a plume is an interesting approach. The authors chose a simplified description of the plume shape to extract quantitative information. This description is again based on poorly justified assumptions. The assumption of a constant wind speed in the lowest 300m of the PBL deserves a more detailed explanation. One would expect that the wind speed increases with height, and that measurements near the ground are not necessarily representative. The estimate of the plume rise velocity should be described in more detail. Mixing and dilution of the plume should also be considered, in particular in the horizontal, since the observations average in the vertical direction. The authors argue that a smaller concentration in the zenith is unreasonable (page 12681, line 10 -12). Depending on the mixing properties of the atmosphere I would not be surprised if the plume is diluted over a time-span of 10 minutes. To overcome the apparent underprediction of the zenith observations of the plume, the authors expand their model to an elliptically shaped plume. While this may indeed be a better description, no explanation is give as to why the zenith observations now yield much higher mixing ratios than those at lower elevations (see Figure 9).

In summary, the manuscript presents an interesting data set of remotely sensed urban NO_2. However, the lack of an error analysis and the missing discussion of the uncertainties associated with the quantitative data presented in the paper needs to be remedied. The assumptions made to interpret the data need to be justified in much more detail or, alternatively, the data needs to be interpreted based on detailed radiative transfer models. In its current form this manuscript does not meet the thorough standards of an ACP publication.

References

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Hönninger G., von Friedeburg C., Platt U. (2004), Multi axis differential optical absorption spectroscopy (MAX-DOAS), *Atmos. Chem. Phys.*, 4, 231-254.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 12671, 2006.