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Atmospheric Chemistry and Physics Discussions



# *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.

**R. J. Leigh et al.**

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The authors would like to thank the referees for their detailed and constructive comments on this paper. Many of the suggestions have been directly implemented, significantly strengthening the arguments made in this work.

All typographical mistakes have been corrected and additional information added to plots etc where requested.

The following changes have been made in response to reviewer's remarks

Referee 2.

*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.*

As per the suggestion of both referees, this facet of the paper has been significantly improved.

*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 NO2 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.*

The subtraction of the zenith slant column measurement from a given time from each off-axis measurement (after it has been analysed with a zenith reference) effectively does just this (equation 1), as the stratospheric component, plus some of the tropospheric component is removed. This is now explained more clearly in the text.

*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.*

As per a similar comment by referee 1, the influence of clouds has been investigated further and found to be relatively small.



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*The choice of a 2 km absorption path in assumption 3 and 4 is arbitrary. Geometrically this would mean that for a 5 deg 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 calculationsĚ*

As per referee 1, the 2 km path length assumption has been replaced by radiative transfer modelling, assuming a 500 m PBL. It is correct that the PBL will change diurnally and annually, however the prevalence of clouds, and lack of absolute radiance information render PBL height-retrieval problematic with the current dataset. Future analysis will improve on this situation, however for this analysis the PBL height is kept at a constant reasonable level, and the aerosol optical depth used in the RT calculations is maintained at 0.1 in the PBL.

*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 NO2 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 wind speed measurement is taken at 10 m height, this extra piece of information has been added to the paper along with mention of the errors introduced by the lack of vertical variation in the assumed wind profile. See comment on plume modelling below:

*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*



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*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).*

Further comment and explanation has been added in the text regarding the increase in derived concentration in Figure 9. We agree that improved input parameters for wind vectors, rise rates, dispersion and dilution would be a useful addition to future exploitations of this technique, particularly when an orthogonal instrument is available. However at present the reconstruction problem is under-constrained with too few measurement parameters available to realistically reproduce reality. Therefore we have maintained the existing simplistic approach and added in additional text to section 3.2 highlighting the uncertainties involved. We believe that this still usefully demonstrates the potential of the technique without introducing unnecessary complexities and uncertainties.

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