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Atmospheric Chemistry and Physics Discussions

Interactive comment on "High spatial resolution measurements of NO_2 applying Topographic Target Light scattering-Differential Optical Absorption Spectroscopy (ToTaL-DOAS)" *by* E. Frins et al.

E. Frins et al.

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Reply to Referee # 2

Interactive comment on "High spatial resolution measurements on..."; By E. Frins et al.

E.Frins et al.

The authors would like to thank the referee for the constructive review. Before we answer in detail the referee's comments and indicate in detail the incorporated changes (see below), we want give a short overview on the major changes in the revised version of the manuscript.

1) We conducted additional measurements with the new technique. We selected the same location and viewing geometries as for the first measurements in order to ensure a consistent interpretation. We made measurements during two days in August and September 2008 covering each a period of about 2-3 hours (the time we could operate the system without external electrical power. The new data sets in general confirm the findings of the initial data set. In addition important information on the internal consistency and the retrieved NO2 mixing ratios from measurements at different targets could be derived. As expected the fluctuation of the results for the closest target were largest confirming the expectations of the effect of the absorption paths above the instrument and target.

2) We added a new section (2.3) on the determination of the measurement uncertainty and detection limit from the DOAS analysis.

3) We added an individual section on the measurement results (new section 3) and we added two new figures showing the results for the additional measurements.

Detailed response:

General Comments: This paper describes application of ToTaL-DOAS to the measurement of atmospheric pollutants, namely NO2, over relatively short distances for high temporal and spatial measurements of pollutants. I would describe this method as a hybrid of classical DOAS (active DOAS), and passive DOAS (utilizing sky scattered sunlight), in the sense that the method does not use an active light source; it depends on scattered sunlight off targets, but it does have a well defined pathlength. The method has the advantage that it could be used in areas with high concentrations of pollutants with minimal infrastructure (no retroreflector, low power use do to absence of active light source), but suffers from the disadvantage that measurements can only be made during periods of adequate solar light. The paper as such, discusses the methodology necessary to make such measurements, and presents a small data set of measurements in an urban area.

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Interactive Comment

Specific Comments: My largest objection I have with the manuscript as presented, is the relatively small (and selective?) data set that has been presented in a paper that has largely been presented as an application paper of a technique that can obtain high temporal resolution data. The small number of data points (8 total) that have been presented are problematic. Three far targets were chosen and the instrument can sequentially switch between the different targets and a zenith measurement (not utilized in the end). And yet the data presented in Figure 4 show only 1 data point each from targets T2 and T3. Certainly a statistical comparison of measurements from T1, T2, T3 or the discussion of comparison between these measurements should have relied on more than 1 measurement from each of the alternate targets.

Author's reply: 1) We agree with the reviewer that the previous manuscript version contained a small data set. In the revised manuscript we have added new measured data (see Fig. 5,6), made under a bit little different weather conditions (e.g., the temperature in October/2008 is lower than in November last year). Especially from these new measurements good consistency of the results for the different targets is found. We think that these findings provide reasonable evidence for the usefulness of the proposed method for measuring trace gas concentrations at short light paths. Note that in the new section on the measurement uncertainties (2.3) we also use the zenith spectrum to estimate possible systematic errors of the spectral analysis.

Section 2 Line 12 - "three small pieces of concrete". The orientation of these reference pieces of brick, with respect to the sun and the far targets, is not described and would be welcome. Are they oriented in the same plane as the walls of the buildings in order to reflect light from sky and sun in the same direction? One brick was chosen? Why? Is there any difference between them?

Author's reply: We added information on the orientation of the small pieces of concrete in section 2.2. and the discussion and conclusions (new section 4). We now explained in detail that the selected piece of (painted) concrete was chosen, because it yielded the smallest residual.

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The distance from instrument to brick is d=0m. Was it placed directly in front of the mini DOAS for the reference measurement, or was the DOAS turned 90o to collect light from it. Not enough experimental details are given.

Author's reply: We did not turn the instrument for the measurement of the reference spectra. This information is now added to the text in section 2.2.

A more explicit description of the color of the reference brick would be useful. Was it painted. Was it the same color as the target walls, etc, etc.

Author's reply: The surface of the used piece of concrete had remnants of white color (like the fat targets). We added this information to section 2.2). Nevertheless, we would expect that there are very probably differences in the spectral reflectances of the different white paints, caused by their composition and age. The systematic investigation of such differences should be the subject of future studies.

Section 2 Line 26 - "Assuming a detection limit of ...." How was this arrived at? One should not assume a detection limit for SCD obtained from an alternate method. The authors have described that the residuals for fits using these artificial targets were very large. The detection limit should be derived from the data generated in this measurement method, which could be much higher. The assumption should be supported.

Author's reply: We added a new sub section (2.2) with a detailed discussion of the errors arising from the DOAS analysis. The reference to the paper by Stutz and Platt is added.

Figure 3 and discussion thereof: The 4 options shown in the figure are simplistic and arbitrary. The underlying assumption is that a well defined emission cloud (plume) exists in a predetermined shape and it is homogeneous. Stating that no error arises for case a) and d) is only true for a homogeneous emission cloud, and if the NO2 concentration outside the cloud is the same at the instrument and the building. In reality the plume will be inhomogeneous. The assumption on line 26 (maximum difference =

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20m) is unsupported, and apparently arbitrary. It is not clear how an error of 13% and 8% are arrived at.

Author's reply: We agree with the referee that the four options shown in Fig. 3 are simplistic and somewhat arbitrary, and that the underlying assumption of our method is that a well defined emission cloud exists in a predetermined shape and it is homogeneous. Of course, in reality the plume will be inhomogeneous, but at present we have no material means to estimate this error through experimental measurements under a variety of measurement conditions. The intended purpose of Fig. 3 was only of didactical character. For the sake of clarify, in the revised manuscript version we skipped some debatable affirmations (e.g., 20m difference and the 13% and 8% errors), as suggested by the referee.

Are we meant to compare these to the actual measurements made on Target 1 and Target 3??

Author's reply: We agree that from our initial data set, no conclusions on the effects of the influence of plume homogeneity could be drawn. In contrast, the new measurements now allow important conclusions on the effects of the homogeneity of the NO2 concentration field (see new section 3), which confirm the expectations from the discussion about the basic effects of the plume shape. In particular it becomes obvious that these effects are strongest for the observations of the closest target.

The authors are making an attempt to communicate a real problem....that errors will arise from inhomogeneities within the measurement region, especially if the solar light radiating the reference material and target material do not encounter the same column of pollutant material on approach. Since the plumes will never be homogeneous and well defined, I believe the only way to estimate this error is through experimental measurement under a variety of measurement conditions (wind directions, using all three targets, possibly with a co-located point source measurement of NOx, etc).

Author's reply: Unfortunately, there are no other methods available for doing compar-

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ative measurements. Despite this, we think that important conclusions can be drawn from the internal consistency of our own observations, especially taking into account the new measurements. This is now discussed in much more detail in the (new) section on results (section 3) and the section on discussion and conclusion. We think that these findings provide reasonable evidence for the usefulness of the proposed method for measuring trace gas concentrations at short light paths. We realize that additional work might be necessary to fully exploit the advantages and drawbacks of ToTaL-DOAS. Despite this, we think the especially with the additional measurements we could demonstrate that the underlying idea is sound. We are confident that our initial work can evolve into a valuable tool for measurements of NO2 and other trace gases, and that the present work is a step in that direction.

Section 3 Line 6. One cannot obtain standard deviations for target 2 and target 3 if only a single measurement is available (as Figure 4 would suggest). Do the authors really mean standard deviation of separate measurements, or do they mean standard error of DOASIS fit? Please clarify. If more measurements are available for T2 and T3, they certainly should be presented.

Author's reply: The error is determined from residual of the spectral DOAS analysis. We added a new section (2.3) with detailed information on the determination of the error.

Technical Corrections: Throughout - please remove the word "basically" throughout the manuscript. It is redundant. Section 2 Line 24 - reword. Was the reference brick also painted white? Figure 2: the figure caption or y-axis labels do not adequately describe what is being shown in the figure. There are two lines for each sub figure, and the composition of each line is not described. The difference between a, b, c components of the figure are not defined adequately; they are not simply expansions of the same data, as I originally thought. Which target, which reference were used to generate the figures? Figure 3. One caption is needed, not two. Caption for 3c has a typo. Should be instrument (S). Figure 4. Uncertainties are not shown for the single points for T2

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and T3. Section 3 Line 25 . Change to "several tens of meters". References - I cannot find where Frins 2006b is referred to in the text??

Author's reply: We performed all minor technical corrections mentioned by the referee. We suppressed the word "basically" throughout the manuscript, and re-worked Figs. 2, 3 and 4.

We hope the revised manuscript version meets the referee's requirements.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 10257, 2008.