Author's Response to Anonymous Review #1 (AMTD, 3, 2317-2366, 2010)

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Introduction

First of all, the authors greatly acknowledge the anonymous reviewer for carefully reading the manuscript and for giving constructive comments and suggestions that have led to clear improvements.

This document contains the author's response to the comments of anonymous reviewer #1. Each comment by the reviewer is discussed separately with the following typesetting: The exact reviewer's comments are in italics (numbered C1, C2, etc.), the author's response is in roman (numbered R1, R2, etc.), and the changes in the manuscript are typesetted in bold.

Review Comments and Author's Response

[C1] *The sensitivity study performed and illustrated e.g. in Table 1 demonstrates that the main source of uncertainty on the* NO_2 *column retrieval is related to the boundary layer height. Why not including this as an additional free parameter in the look up table? One may easily think of a modified retrieval algorithm where the BLH could be iteratively optimized until best agreement between columns retrieved from the 3 angles is achieved. Is there a technical limitation to this approach, please comment on this possibility.*

[R1] As the reviewer remarks, the boundary layer height (BLH) is the main source of uncertainty. The suggested possibility of including the BLH as a free parameter was also considered by the authors as a possible extension of the two-step algorithm.

Including the BLH as a free parameter would be especially relevant in the second step of the algorithm, after the AOT retrieval based on relative intensity, which is quite unsensitive to BLH. There is no specific limitation to apply BLH as a free parameter, and to find the specific BLH where the agreement between columns retrieved from the 3 angles is largest (smallest spread).

However, the reason not to choose this approach was two-fold. Firstly: Improving the algorithm by a good BLH estimate is part of future work, driven by the conclusion of the present paper – i.e. the uncertainty in the tropospheric NO_2 column retrieval due to the unknown BLH. This improved algorithm is still under development. The main source of information for an estimate of the BLH will be MAX-DOAS observations of O_4 absorption (together with relative intensity observations), although NO_2 will also contribute to the BLH estimate. Several questions are currently still open.

Secondly: Including BLH as a free parameter would possibly lead to misattributing the real origin of the spread ε (as defined on page 2336, eq. 17) between the three elevations (α=4°, 8° and 16°) to a false BLH. This spread may have many origins (see p. 2337, l. 3-6), only one of which is a deviation from the assumed BLH.

Since the distribution of NO_2 and aerosol in the atmosphere may be much more complicated than assumed in the model (both have their own horizontal and vertical distribution in the real atmosphere), a simultaneous change of the NO_2 and aerosol layer heights in the algorithm (the free BLH parameter) is only one of multiple free parameters one could think of. E.g. another possibilities for free paramters would be: to separate the NO_2 profile height from the aerosol profile height (and use only one of the two as a free parameter), the profile shape, or aerosol microphysics. This could also reduce the spread.

In our view it would lead to an ambiguous interpretation of the error estimate (the spread as defined on page 2336, eq. 17) if only one of the various parameters leading to uncertainties would be singled out.

Including the BLH as a free parameter would complicate the retrieval scheme, whereas the strength of the present approach lies in the simplicity and straightforward interpretation of both the retrieved vertical NO_2 column and its error estimate. The BLH as a free parameter would in many cases not be the real boundary layer height due to the interference with other parameters.

The following sentence has been added to the manuscript:

- (p. 2337, l.7) **Since the boundary layer height may well be the parameter with the largest contribution to the uncertainty ε in the tropospheric NO_2 column retrieval (see Table 1), the two-step algorithm could be modified by including the boundary layer height as a free parameter, changing it iteratively, by minimizing ε. However it was decided in this work not to apply this additional step, as the boundary layer height is not the only parameter affecting ε, as described above.**

[C2] *p.2325, l.16: Note that the approach of subtracting the signal from the blind pixels below the ozone cut-off not only correct for the read-out offset, but also for the dark current itself, which added to the fact that integration time for* *individual acquisitions is generally small (of the order of 1 sec or less) probably explains that a specific dark current correction is not necessary.*

[R2] This suggestion of the reviewer, it will be included in section 2.3.1:

- (p.2326, L3) **Tests on temperature-controlled measurements, under representative measurement conditions, have shown that differences between including and not including DC-correction was less than 0.1% in the fitted NO_2 differential slant column. A possible explanation for this small effect is that the integration time for individual acquisitions was generally small (of the order of 1 s or less), resulting in a low DC, and that the EO correction described here also includes a correction for the average DC. Only the pixel-to-pixel variations on top of this average DC are not accounted for. For other trace gases with smaller tropospheric column amounts the effect of not correcting for DC will be larger.**

[C3] *p.2326, l.10: why not interpolating zenith-sky measurements at the time of the off-axis measurements. In particular for near-twilight measurements, this would allow for some compensation of the fast changing light path and possibly photochemical state.*

[R3] The authors agree with the reviewer that interpolation of the zenith-sky measurements to the time of the off-axis measurements will improve the retrieval, especially of near-twilight measurements.

This approach was not applied for practical reasons. The main reason was that each off-axis spectrum is analyzed in our approach with the nearest-in-time zenith spectrum. By application of this method, only differential slant columns for the off-axis elevations can be retrieved, and not for the zenith, since the zenith spectra cannot be their own reference. As a consequence, there are no zenith-sky differential slant columns to interpolate on after the DOAS analysis.

There were several reasons for the authors to choose the nearest-in-time reference method: (1) this makes the DOAS analysis less sensitive to instabilities of the instrument (e.g. thermal instabilities) since each spectrum is analyzed with a reference that was taken under almost the same conditions, (2) this approach makes the analysis of all off-axis spectra of one day less sensitive to one and the same reference, which is seen as an advantage since it makes the differential slant column observations more independent of other observations taken at different times of the day, (3) the nearest-in-time reference method was chosen in combination with a short integration time of 30 seconds per elevation in order to achieve a high temporal resolution. The authors were aware of the fact that this relative short integration time leads to qualitatively poor spectra (low signal to noise) around sunrise and sunset, and to errors in the retrieval due to a semi-simultaneous zenith reference which leads to a stratospheric contribution to the NO_2 differential slant column observation. This was, however, considered to be a minor problem given that the focus of research was not on this period of the day, but rather on the middle of the day (which is the time of satellite overpasses).

The following lines will be added/changed to the manuscript:

- (p.2326, l.13) **Since spectra were measured within 30 s to 2 min from the zenith measurement, the change in stratospheric path length was of relatively little influence, except around sunrise and sunset.**

  Simulations were performed to study the error introduced by using a semi-simultaneous reference spectrum, as a function of the solar zenith angle. Here equal NO_2 column amounts were assumed for the stratosphere and the troposphere, and no temporal dependence. The error in the NO_2 differential slant column is below 1% for solar zenith angles smaller than $74°$, and below 5% for solar zenith angles below $82°$. For a representative day in March (around the equinox), this implies that 8.5 out of 12 hours of daylight have an error below 1%, and 10.7 hours have an error below 5%.

- (p.2335, l.17) **Systematic error contributions to the observational error are: (1) errors caused by incorrect knowledge of the actual field-of-view, which may be caused by incorrect aiming of the instrument (e.g. when it is unattended after periods of heavy winds) or by imprecise knowledge of the offset in the field-of-view as described in Sect. 2.1, (2) incorrect electronic offset correction of raw spectra, (3) errors in the differential cross-sections of NO_2 (e.g. temperature dependency), (4) errors due to the use of a semi-simultaneous reference spectrum (see Sect. 2.3.2).**

[C4] *p.2328, l.8: Is the earth curvature really the limiting factor here? I would expect that the assumption of homogeneity in the BL is maybe causing a larger problem than earth curvature for low elevations.*

[R4] The authors agree with the reviewer that the earth curvature is not the only limiting factor. In addition to the reason mentioned by the reviewer, namely the assumption of homogeneity in the BL, another reason not to include the elevation is the relatively large effect of small misalignments at this low elevation. The authors propose to rephrase l.8. to:

- **Smaller elevations were not used for several reasons. Firstly, because inhomogeneities in the distribution of aerosols and NO_2 in the boundary layer, which are not included in the forward**

radiative transfer modeling, will have a larger effect at smaller elevations and therefore lead to larger uncertainties in the retrieval. Secondly, because small misalignments will also have the largest effect for the smallest elevations. Thirdly, because the effect of the curvature of the Earth is not captured by the radiative transfer model, which will only have a noticeable effect in the VIS, for small elevations under very clear conditions (see also Sect. 3.1.1).

[C5] *p.2334, l.16: It is correct that intensity measurements are weakly sensitive to changes in the BLH (this is clear from table 1), however it should be mentioned that intensities are in contrast very sensitive to the presence of clouds. In fact more than* O_4 *observations.*

[R5] The authors agree that relative intensity measurements are very sensitive to the presence of clouds. This is also explicitly mentioned in e.g. Section 4.1 ('The first day .... give similar AOT values.') and in the conclusions (p.2343, l.25). The line referred to by the reviewer will be changed to:

- **The relatively weak sensitivity of relative intensity to a change in boundary layer height, is the reason that relative intensity observations are more suitable for boundary layer aerosol optical thickness estimations than measurements of O_4, but only in the absence of clouds.**