Interactive comment on “Determination of tropospheric vertical columns of NO₂ and aerosol optical properties in a rural setting using MAX-DOAS” by J. D. Halla et al.

Anonymous Referee #2

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Review of the paper “Determination of tropospheric vertical columns of NO₂ and aerosol optical properties in a rural setting using MAX-DOAS” by J. D. Halla et al.

In this manuscript, results from three weeks of measurements with a MAX-DOAS instrument during the BAQS-Met field study are reported. The instruments and methods used are described, comparisons with other data sets are presented and several case studies are discussed. The paper is overall well written but in parts is too detailed. It reports on interesting measurements and comparisons and contributes to the overall description of the situations during the BAQS-Met field study. I therefore think it could be published in the BAQS special issue but only after major revisions as discussed
I have several concerns about this paper:

1) A large part of the manuscript describes the DOAS retrievals and their inversion to vertical columns and aerosol optical depth. However, it is not clear which part of this is really a new development and which is just application of previously developed concepts. My impression is, that the new aspect in this study is the application of the method to three weeks of data and that no really new concepts are presented. I therefore think that this part should be shortened considerably. At the same time, it has also to become clearer as some important aspects remain confusing to the reader, for example if a fixed FRS background was used as stated in the text or if the closest zenith-sky measurement was taken as most of the discussion implies. I’m also surprised that the quantities H_NO2 and H_aerosol which are introduced in the text are never used later although this would be quite interesting (see detailed comments below).

2) The comparison of the retrieved NO2 VC with satellite data and profiles derived from air-borne observations is discussed in some detail. While the approach taken is sound, the results are very much limited by the small number of coincidences. For SCIAMACHY, only one comparison was possible and even for OMI, only 8 points remained! I don’t think that this is contributing significantly to the validation of these data sets, and discussion of the differences to results from previous comparisons is of very limited interest.

3) The air-borne profiles used for validation have problems as well – apart from the fact, that they were taken at different distances from the MAX-DOAS instrument for different altitudes and do not cover the interesting altitude region, they also are identical within their scatter for the two profiles shown in Fig. 8. Still, the authors construct two different profiles from these data which I think is not supported by the measurements.

4) In the last part of the manuscript, several case studies are discussed, highlighting
the usefulness of MAX-DOAS observations as compared to in-situ or active DOAS measurements. While the study on June 30 is quite straightforward, I do have problems with the vertical NO2 columns presented for July 2nd and July 9. In both cases, large NO2 columns are derived in times with little separation of the viewing directions. In particular on July 2nd, a rather small change in DSCD_30 (factor of 2) leads to a large change in VCD (factor of 5). In this situation, the DAMF_xx appears to be only of the order of 0.2 for all directions, indicating that the sensitivity of the measurements to the NO2 is very small. How realistic are then the results? Can the enhancement in NO2 column also be seen in the zenith-sky observations? Could this possibly also be related to the aerosol layer mentioned in the text? I’m also worried by the fact that the AOD is actually reduced during these episodes – if you could see the haze from the fires, AOD should certainly be significantly enhanced!

Details comments:

P 13037, L4: have been estimated

P 13037, L25: not sure the size of the molecules is the right quantity here – H2 is small but cannot be measured by DOAS while O3 can.

P 13038, first para: This paragraph is a mix of different things and should be rewritten

P13038, l17: unlike for active DOAS

P13041, l28: Here, it is stated that a single FRS was used for all retrievals. Later, this does not appear to be the case. What has been done in the study? What is shown in the figures?

P13043, l15 and P13044, l11: no need to reference DOAS again and again

P13043, l26. How do the assumptions on the NO2 profile made for the SCIAMACHY retrieval fit to your airborne profiles from Fig. 8?

P13047, l22 and 13059, l11: I don’t see the relevance of the Friess et al. paper here
as it does not deal with NO2 retrievals or the criterion used for the geometrical approximation

P13048, l23: most OR predominantly

P13049, l7: why top of the troposphere and not top of the atmosphere?

P13049, l20: How was the fact that you derive aerosol properties at 360 nm but retrieve NO2 around 420 nm be treated in the retrieval?

P13050, l20: I assume that SCD_90 was subtracted from SCD_x, not the other way round

Section 4.2: Why did you not compare the air-borne NO2 profile with H_NO2 derived from the retrieval?

Conclusions: First sentence: As stated above, I don’t see that you outline a new method in this paper. As it is not new, it should be described only very briefly.

Conclusions: Comparison of VC_RTM and VC_geom – you state that the agreement is relatively good but only for those cases where the quality criterion is fulfilled. What about the other cases – how does VC_geom_30 compare to your VC_RTM then?

Table 3: It would be very interesting to add here the meteorological boundary layer height as well as H_NO2 as derived from the retrieval

Figure 3: The box DAMFS O4 in the “Field Measurements” doesn’t make any sense – this should come from the RTM branch above

Fig. 8: Please use the same scale for both figures. It will then become obvious that the two aircraft profiles are identical within the scatter of values. I don’t see good arguments from the data to derive separate NO2 profiles from them apart from the active DOAS point on the ground.

Fig. 12: Why are there so few NO2 VC values? There are more AOD values which I
find odd.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 13035, 2011.