

Interactive comment on “Near-surface and path-averaged mixing ratios of NO₂ derived from car DOAS zenith-sky and tower DOAS off-axis measurements in Vienna: a case study” by Stefan F. Schreier et al.

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We would like to thank the reviewer for his / her useful comments.

General Comments: This paper presents near-surface and path-averaged mixing ratios of NO₂ derived from car DOAS zenith-sky and tower DOAS off-axis, measurements performed in Vienna city during several days on 2015 and 2016. This paper provides an useful intercomparison between tower DOAS, mobile DOAS and in-situ observations. Specific Comments: Section 2 - Instrument and car journeys, in this section you should

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add few info about the in-situ instruments (type, error, etc.). Also please add a map (a new Figure) or include in Figure 1 the location of the in-situ monitoring stations and also the location of the DOAS tower instrument. We have now rewritten the first passage of Sect. 3.3 and added information about instrument type and error of in situ instruments. In addition, we now refer to a recently published report (Spangl, 2017) (see Page 14, Line12-20). In this report, which is available online, all the air quality monitoring stations are described in detail (e.g. instrument type, location, surrounding, etc.).

We have now indicated all in situ stations in Figure 1 that are used in combination with the car DOAS measurements, but also those that are used for comparison with tower DOAS measurements. We have now renounced to include the Table with all the station name/coordinates as this information about the in situ stations as even more details can be found in the mentioned report anyway. Also now shown in Figure 1 is the position of the Danube Tower, on which tower DOAS measurements were performed. We have now included a sentence to describe what is seen in addition to the exemplary car route in this new Fig. 1 (see Page 8, Line 1-4).

Please describe the tower DOAS instrument, I suggest you to introduce a Table with the technical characteristics of the two instruments (tower DOAS and mobile DOAS).

The one and the same DOAS instrument, which we used for both car DOAS and tower DOAS applications, is already described in the version of the manuscript (see Sect. 2.1, 2.2, and 2.3 of the ACPD Manuscript Version). In order to provide more technical details of the DOAS instrument, we have now introduced a Table with the technical characteristics (see Table 1).

Figure 2. Could you explain the very low peak of intensity? Is it related to a tree, tunnel, or a bridge? Did you filter all the DSCDs function of RMS and O4?

The very low peak of intensity in Figure 2 is related to the tower DOAS measurements and shows up once every rotation of the tower platform, e.g. when the DC Tower (a

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skyscraper), which is about 1 km away from the Danube Tower, blocks the field of view of the instrument. We have already described the reason for this low peak of intensity in the manuscript and used this peak for determining the exact orientation of the tower platform (see Page 8, Line 15-23 of the ACPD Manuscript Version). For the car DOAS zenith-sky measurements, we filtered all the DSCDs as a function of chisquare, e.g. NO₂ DSCDs with chisquare values > 0.025 were not included in the analysis. This filtering is already described in the manuscript (see Page 9, Line 17-18 of the ACPD Manuscript Version). For the tower DOAS off-axis measurements, we did not apply any filtering. We have now added a sentence mentioning that tower DOAS off-axis measurements are not filtered (see Page 10, Line 5-7).

Figure 3, please introduce the DSCD error. Also please introduce the error of each DSCD presented in the manuscript.

We have introduced and evaluated the error for each NO₂ DSCD presented in the manuscript. In general, the error of (unfiltered) DSCDs is lower than 0.75×10^{15} molec cm⁻² for car DOAS zenith-sky NO₂ DSCDs and lower than 1.5×10^{15} molec cm⁻² for tower DOAS off-axis NO₂ DSCDs presented in the manuscript (see Figures in the supplement). We have now added a sentence to give an overall (maximum) error of NO₂ DSCDs for both car DOAS zenith-sky and tower DOAS off-axis measurements (see Page 10, Line 8-10).

3.2.1 Temporal resolution and computation of horizontal NO₂ gradients- Could you specify the exposure time for the mobile DOAS instrument? (or this info could be included on the suggested Table for the two DOAS instrument).

Typical values of the exposure time for car DOAS zenith-sky measurements were generally between 0.00625 and 0.1 seconds. In most cases, however, the exposure time was 0.025 seconds. We have now added a sentence to specify the exposure time for the car DOAS zenith-sky measurements (see Page 10, Line 14-15 and also added this information in the new/additional Table 1.

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3.2.2 Stratospheric NO₂ columns, Could you specify the error of Bremen 3d CTM (B3dCTM) model?

It is difficult to quantify the accuracy of the stratospheric NO₂ columns from the Bremen 3d CTM. In absolute units, it is not very good as it is a free running model without data assimilation. However, in this analysis, the stratospheric model is only used for the diurnal cycle of the stratospheric NO₂ column as the absolute value is scaled to GOME2 satellite observations at the time of overpass. The uncertainty of the diurnal variation is large at twilight but small during the day as changes in stratospheric NO₂ are small when compared to tropospheric NO₂ columns in polluted regions. As a rough estimate, the uncertainty of the stratospheric correction is assumed to be less than 10% or typically 1×10^{15} molec cm⁻². We have now added a sentence to highlight the uncertainty in the stratospheric correction (see Page 12, Line 1-6).

3.2.3 Conversion to tropospheric NO₂ vertical column densities SCD_{ref}, could you specify why you don't have a SCD_{ref} for each day? SCD_{ref} is quite important if you want to have qualitative data. I suggest to the authors to introduce more details about SCD_{ref} calculation, e.g. exact time of the selected SCD_{ref}. SCD_{ref} having 1.3×10^{15} , 1.1×10^{15} , and 2.2×10^{15} molecules/cm² as tropospheric contribution could be realistic. Considering that SCD_{ref} contain stratospheric and tropospheric contributions, did you cancel the stratospheric contribution? why do you refer to SCD_{ref} as having only tropospheric contributions?

We agree that it is important to have as many as possible SCD_{ref} measurements for quantitative data analysis. The reason why we didn't use SCD_{ref} of each single day in our study is that for most of the days, (noontime) SCD_{ref} was taken in urban areas, where pollution levels are expected to be higher. The three SCD_{ref} measurements that we used were recorded during noontime and outside of Vienna in rather rural areas, where pollution levels are expected to be low. According to Wagner et al. (2010), AMT, spatially inhomogeneous tropospheric trace gas concentrations are a prerequisite for the "zenith-sky only" approach to avoid large systematic errors when applying Eq. 11

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in their paper (Eq. 1 in our ACPD Manuscript). By using SCDref measurements in areas with rather small tropospheric NO₂, as we did in our study, the errors are kept as low as possible. The selection of the three SCDref measurements was a compromise between having such measurements during noontime in unpolluted regions and at the same time to keep the time difference between SCDref and the days for which these SCDref measurements are used as low as possible (< 9 days). There are other examples in the literature where only few SCDref measurements are used for similar DOAS-type analyses (e.g. Tack et al. 2015, AMT). The authors of that study use a single SCDref measurement for a period of about 40 days. Because a similar approach to convert zenith-sky DOAS measurements into tropospheric NO₂ vertical columns is described in the latter publication, we have now included this study in the references. We have specified the exact time and also the location (lat/long) of the three SCDref measurements that we used for our data analysis in Table 2. In addition, we have now added the exact time, SZA, and location of the three SCDref measurements used in our study and also we have now added more infos about how we calculated SCDref in Sec. 3.2.4 (Page 13, Line 12-21). In order to not confuse the reader, we have now avoided the use of “tropospheric” amounts in SCDref and replaced it with “residual” amounts in Sect. 3.2.4 and 3.3. This formulation was already used in the literature before (e.g. Tack et al., 2015, AMT).

A chapter to describe the AMF calculation (using NO₂ profiles, albedo, geometry, PBL, etc.) is mandatory for this study, I suggest to the authors to use a table. Figure 6 should be part of this section and should include the AMF calculations for several days which are presented in this study.

In response to the comments of both reviewers, we have re-evaluated the AMFs used in the study by adding a sensitivity study of AMF changes for realistic values of AOD, single scattering albedo and mixing height in Vienna. Based on the results we have decided to change the AMF used to values based on an intermediate scenario which according to our sensitivity study provides a good compromise. All other scenarios are

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well within 20% of these values. Therefore, we have re-calculated VCDtropo NO₂ in our study and plotted the respective figures again. Both the sensitivity study and the description of the AMF used have been included in a new Section (Sect. 3.2.3, Page 12, Line 8-27) in the revised manuscript.

The authors should give more details about the error calculation of tropospheric NO₂ VCD, or a section of errors would be more appropriate.

Uncertainties in tropospheric VCDs are introduced by uncertainties in the quantities used in equation 1 of the manuscript. Assuming that the stratospheric AMF is well known, the uncertainties of DSCDmeas, SCDref, SCDstrato and AMFtropo need to be considered: (see Equation in the supplement) an overall uncertainty of 25% is found, dominated by the assumed 20% uncertainty of the AMF. For situations approaching twilight, the absolute uncertainty of the stratospheric correction increases, and the relative uncertainty of the slant column can become the dominating error source. If the background measurement SCDref cannot be taken in a clean region, then the absolute uncertainty on this quantity can become large and important for the overall uncertainty (see Wagner et al., 2010). We have now added a paragraph in order to provide information on the overall uncertainty of VCDtropo in our study (see Page 13, Line 27-28; Page 14, Line 1-9).

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