General comments

The manuscript entitled 'Horizontal distribution of tropospheric NO2 and aerosols derived by dual-scan multi-wavelength MAX-DOAS measurements in Uccle, Belgium' by Dimitropoulou et al. describes a novel method for the retrieval of information on the horizontal distribution of trace gases and aerosols from MAX-DOAS measurements by exploiting the wavelength dependence of the light path length of scattered sunlight. I was asked by the Associate Editor after the open discussion phase to review the paper and these are my first comments to the manuscript. In the following, line and page numbers are referring to the manuscript version with tracked changes (amt-2021-308-ATC1.pdf).

So far, retrievals of information on the spatial distribution of trace gases and aerosols from MAX-DOAS measurements were mostly restricted to the vertical dimension. Based on the wavelength dependence of the light path through the atmosphere, the novel method proposed here enables to gain also information on the horizontal distribution, which is a great benefit, in particular if the spatial distribution of trace gases is highly variable, such as traffic emissions in urban areas. Therefore, the manuscript fits well in the scope of AMT.

A main problem of the manuscript is that the description of the methods is lacking conciseness, and it is not always clear whether certain approaches are based on physical principles or rather represent simplifying assumptions. In particular, the description of the horizontal distribution inversion approach (Section 4.3.) conveys the impression that a constant weighting function as a function of distance with a sharp drop at the estimated light path length is a fact, while this is in actually a very simplifying and physically incorrect assumption. In reality, I would rather expect that the sensitivity decreases exponentially with distance from the observer in accordance with the Beer-Lambert law, as already discussed by Kern et al. [2010] and Vogel et al. [2011]. In the light of these (over-) simplifications, it is surprising to see that the retrieved horizontal distribution compares very well with airborne and satellite measurements.

I wonder if it is not possible to infer the correction factor discussed in Section 4.2 directly from the aerosol and NO2 profiles retrieved by MMF, which directly provide dSCD(NO2) and c(NO2) and thus L(NO2) as the ratio of both.

Some of the measurements used for intercomparison are not explained, or only later in the manuscript. For example, it is not clear what CIMEL measurements are, and airborne measurements are mentioned already in Section 4.3, but the context is only given much later in the manuscript in Section 5.2. I suggest to add a short Section (4) with a short description of the ancillary data used for intercomparison (CIMEL, APEX, car DOAS, etc.).

The level of agreement between the horizontal distributions of NO2 from MAX-DOAS on the one hand and from satellite, car- and airborne measurements on the other hand presented in Section 5.2. is quite impressive. However, I recommend a major revision of the manuscript due to significant deficiencies in the methodology and in the description of the inversion approach. The inversions of the horizontal distribution need to be re-done using physically correct weighting functions.

Specific Comments

Section 4.3: I find the description of the forward model quite confusing, and I feel that this Section requires substantial revision. Moreover, I think the inversion needs to be re-done with appropriate weighting functions. Equation 12 suggests that the forward model for the calculation of a mean concentration is given as the integral over the concentration divided by the light path length. First of all, I guess that the model is not based on numerical integration, but that it is rather based on a discrete sum over the horizontal grid. Second, what is missing in this equation is an appropriate weight as provided by a weighting function that represent correct physics. The assumption of a constant sensitivity between the instrument and the effective light path length L is not realistic. Instead, an exponential decrease should be chosen as weighting function in accordance with the Beer-Lambert law. Also, the light path through the boxes is not horizontal but slanted, so the weighting function needs to contain the cosine of the elevation angle (this is however only a small effect at 2° elevation angle). I furthermore think it would be more appropriate to use the observed dSCDs directly as measurement vector ***y*** instead of (weighted) mean concentrations, which are not a very useful quantity. The weighting function as a function of wavelength λ and distance x to the box would then be

$$K(\lambda, x) = (1 - \exp(1\text{-x}/\mathrm{L}(\lambda))) * \Delta x/\cos(\alpha)$$

with Δx being the width of the boxes, α the elevation angle, and $\mathrm{L}(\lambda)$ the effective light path estimated from the measurements. The forward model would then simply be

$$SCD(\lambda) = \sum_i K(\lambda, x_i) * c_i$$

with x_i being the distance between observer and box i and c_i the NO2 concentration in this box. This equation can be readily inverted using OEM.

P29, L527ff: It cannot be 'seen' from Figure 11 that the weighting functions are constant up to a distance L, but instead this is an assumption, which is physically incorrect – see my comments above.

P29, L552: Here you mention CIMEL observations without explaining what the nature of these measurements are and where they have been perfromed. A short description of these measurements should be part of an extra Section on ancillary measurements further up in the manuscript – see general comments.

P29, L550ff: Do I understand it right that the linear decrease in a priori AOD is a decrease in the horizontal dimension? If so, what are the motivations for this assumption? A higher AOD at your measurement site than anywhere else in the surroundings is hard to justify (except if there were strong aerosol sources next to your instrument).

P30, L567: What kind of airborne observations are these? This is only explained later in the manuscript – I suggest to move the introduction of APEX from Section 5.2. to a Section further up in the manuscript describing all ancillary data used in this study (see general comments).

P30, L580ff, and Section 4.4: It is stated several times that there is no information on the horizontal distribution at distances closer than the shortest scattering distance. The averaging kernels are, however, not zero at these regions (see revised Fig. 15). Instead, it seems that parts of the information coming from short distances are falsely attributed to distances further away. For example, the 8.75 km averaging kernel has a constant value of 0.04 up to a distance of approx. 8 km. Unfortunately, no averaging kernels for distances closer to the instrument are shown.

Figure 15: Given the small peak values of the averaging kernels (at most 0.05), I wonder if the fine horizontal grid of 500 m is really useful or if a coarser grid would have been more appropriate. Furthermore, I could imagine that the averaging kernels would look more smoothly if more realistic (exponentially decreasing) weighting functions rather than the arbitrary step-like functions would have been used.

Section 5.3.1: Here you describe the methodology for comparison between the different datasets, but the NO2 gas maps based this method have already been shown in Section 5.2, if I understand it right. Is there something different in the data processing (filtering and spatio-temporal binning) for the production of Figure 21 compared to Figure 19 (except that Fig. 21 shows seasonal averages)? If not, then the description of this method should appear at the beginning of Section 5.2.

Technical Corrections

P6, L133: The O4 cross sections by Finkenzeller have recently been published [Finkenzeller and Volkamer., 2022]. Please add the according reference.

P8, L185: 'in six different fitting windows' - > 'in the six different fitting windows listed in Section 2.2.'

P9, L187: Explain abbreviations/acronyms 'OEM' and 'MMF'

P9, L189: Explain abbreviation 'MLH'

P9, L195: Here you should state that the NO2 near-surface concentrations and VCDs and the near-surface aerosol extinction are retrieved as a function of distance from the instrument.

P9, L197: Three times 'horizontal' in one sentence. Please rephrase.

Section 4.2: Equation (3) is just a trivial rearrangement of Equation (2) and therefore obsolete. Please remove one of these.

P16, L348: 'Regarding the aerosols' - > 'Regarding aerosols'

P23, L434: I suggest replacing the term 'sanity' with 'consistency'

Equation 14: I suggest to replace dx with Δx since dx can be confused with the differential in Equation 11.

P30, L579: Insert a comma before the year number.

P40, L670: Replace 'error' with 'covariance matrix', and mention that the error is given as the square root of its diagonal elements.

Caption of Figure 16: 'measurement error' → 'measurement and smoothing error'

Figure 18: The bottom-right panel should have the same width as the other panels.

Title of Section 5.3: 'Comparison between MAX-DOAS horizontal NO2 distribution and TROPOMI observations'

P70, L1105: The approach is better than what?

P70, L1108: Do you mean a slope closer to unity?

P70, L1114: Delete this paragraph on the role of the a priori as it refers to a Section that has been removed in the revised manuscript.

References

Finkenzeller, H. and Volkamer, R.: O2–O2 CIA in the gas phase: Cross-section of weak bands, and continuum absorption between 297–500 nm, 279, 108063, https://doi.org/https://doi.org/10.1016/j.jqsrt.2021.108063, 2022.

Kern, C., Deutschmann, T., Vogel, L., Wöhrbach, M., Wagner, T., and Platt, U.: Radiative transfer corrections for accurate spectroscopic measurements of volcanic gas emissions, 72, 233–247, https://doi.org/10.1007/s00445-009-0313-7, 2010.

Vogel, L., Galle, B., Kern, C., Delgado Granados, H., Conde, V., Norman, P., Arellano, S., Landgren, O., Lübcke, P., Alvarez Nieves, J. M., Cárdenas Gonzáles, L., and Platt, U.: Early in-flight detection of SO_2 via Differential Optical Absorption Spectroscopy: a feasible aviation safety measure to prevent potential encounters with volcanic plumes, 4, 1785–1804, https://doi.org/10.5194/amt-4-1785-2011, 2011.