Three-dimensional radiative transfer effects on airborne, satellite and ground-based trace gas remote sensing

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Response to the Reviewer's Comments

We thank the two reviewers for their positive comments, critical assessment and useful points to improve the quality of our paper. In the following we address their concerns point by point. Changes in the paper are shown in blue. We hope we clarified all concerns and that the revised manuscript has improved.

Reviewer 1

Reviewer Point P 1.1 — On p.3, l.43 the authors state that the assumption of horizontal homogeneity (1D-BOX AMF) is not valid in polluted environments. In theory, I fully agree with this statement. However, in case of real-world observations I'm doubting that all relevant data will be available for most regions in order to fully benefit of the 3D-BOX AMF calculations. It requires accurate and high-resolution 3D trace gas and aerosol fields, while it is now often already difficult to get the proper high-resolution a priori for the 1D layer geometry. This has not been discussed in the paper. It would be an added-value to add a paragraph, e.g. in the conclusion, to discuss what is currently missing or needed in order to fully benefit of the 3D BOX-AMF geometry in case of real-world observations.

Reply: We agree with the reviewer that taking full advantage of 3D-box AMFs requires reliable 3D information that is generally difficult to obtain. We will address this question in further studies, where we plan to use city-scale dispersion models to obtain the 3D fields. We added a paragraph in the conclusion on the particular need of 3D box-air mass factors calculations over a polluted area.

> To fully benefit of 3D-box AMFs, 3D radiative transfer calculations require high-resolution 3D distributions of trace gases and aerosols to calculate the total AMF. Such fields are generally difficult to obtain. In a follow-up study we plan to use 3D NO_2 fields from a building-resolving urban air quality model (Berchet et al., 2017) with a detailed representation of both near-surface and elevated (stack) emission sources to further analyze the added value of 3D-box AMFs.

Reviewer Point P 1.2 — I assume it is most likely subject of a future study but I'm also strongly interested to see the impact on total AMFs and VCDs when using 3D BOX-AMFs instead of 'typical" 1D layer AMFs in case of real-world MAX-DOAS and airborne/spaceborne observations. In case this is subject of future work, it could be mentioned in the conclusion.

Reply: We also think impact on total AMFs and VCDs an important point. The effects of using 3D-box AMFs on total AMFs and on VCDs and the horizontal smoothing observed with 3D-box AMFs will be addressed in further studies as described in Point P.1.1.

Reviewer Point P 1.3 — A quantification of the horizontal smoothing of the plume due to geometric effects when 1-d layer AMFs are used would be an added-value in addition to Fig. 10 and 11. For example, a table could be added quantifying the horizontal smoothing for different scenarios of SZA, VZA, plume size and plume altitude.

Reply: Since horizontal smoothing is an important, but also quite complex topic, we plan to address it in a follow-up study in much more detail than possible here. We added a sentence to the conclusion.

> This finding is particularly relevant for ground-based and airborne remote sensing in cities, where considering 3D effects is likely indispensable to reduce systematic errors. This will be addressed in a follow-up study where also the potential impact of 3D radiative transfer effects on the horizontal smoothing of the retrieved trace gas fields will be studied.

Reviewer Point P 1.4 — The reader might also be interested to get some info on absolute computation time and difference between MYSTIC 1D layer AMF and MYSTIC 3D box AMF computation time for a typical satellite or airborne scene.

Reply: We considered that point and added the following paragraph to section 5.1

> The computational cost of calculating 3D-box AMFs is considerably larger than for 1D-layer AMFs. The computational time for calculating 3D-box AMFs for the scenarios here (see Table 1 with SZA=20°, SAA=90°, VAA=90° and VZA=2°) is around 218 seconds with 1 million photons using a single core of our local machine (Intel Xeon W-2175 CPU @ 2.5 GHz). The computational time for the corresponding 1D-layer AMFs is only about 4 seconds with 1 million photons. Note, however, that even less photons would be sufficient to obtain a similar noise level as for the 3D-box AMFs.

Reviewer Point P 1.5 — p.2, l.35 I understand you want to make a distinction between layer-AMFs (1D) and BOX-AMFs (3D). However, in past studies eg Wagner et al. 2007, "BOX-AMFs" were used for what is defined in this work as layer-AMFs. I would add a short statement to clarify. I noticed that you clarify this later on in p.5. I propose to switch it to the introduction.

Reply: We moved the clarifying comment to the introduction.

> Notice that in previous studies (e.g. Rozanov and Rozanov, 2010) 1D-layer AMF were some times referred to as box AMFs. In this study, we will use the terms 1D-layer and 3D-box AMFs to clearly distinguish between them.

Reviewer Point P 1.6 — p.3, l.68: SCD and VCD acronyms were already defined in the introduction. No need to do it here again. There is some repetition here as well like explaining again what a VCD and AMF is. I suggest to remove it from the introduction or remove it here.

Reply: We deleted the repetitions and replaced the acronym definitions by the acronyms in the method section.

Reviewer Point P 1.7 — In Fig. 8b I would expect the two SCD maxima to be east and west of the true VCD at first glance (also when looking at Fig. 9). However, they both seem to be east of the true VCD, with the most western SCD maximum falling together with the VCD maximum. Or do you assume VZA is 0°? In that case it isn't consistent with the example in Fig. 7. Could you please clarify?

Reply: The example shown in Figure 7 uses a different observation geometry than the one used in Figure 8. We updated Figure 7 to show an example from Figure 8 now. We also modified Figure 9 to match the explanation.

Reviewer Point P 1.8 — p.1, l.6: MYSTIC acronym stands for . . . ?

Reply: MYSTIC stands for Monte carlo code for the phYSically correct Tracing of photons In Cloudy atmospheres. We added this in the abstract.

> ..., we implemented 1D-layer and 3D-box AMFs into the Monte carlo code for the phYSically correct Tracing of photons In Cloudy atmospheres (MYSTIC), a solver of the libRadtran radiative transfer model (RTM).

Reviewer Point P 1.9 — p.2, l.45: I would add "at high resolution" after "trace gas remote sensing"

Reply: We modified the text accordingly.

Reviewer Point P 1.10 — p.3, l.63: It depends also on the molecule and aerosol properties (e.g. SSA)

Reply: We modified the sentence as follows:

> Atmospheric scattering and absorption is determined by the distribution and properties of molecules, aerosols and clouds, and depends on the wavelength of the radiation. Molecular scattering is particularly important in the UV range of the spectrum.

Reviewer Point P 1.11 — p.3, l.75: remove "and" after VCD

Reply: We corrected the typo.

Reviewer Point P 1.12 — p.5, l.87: an aircraft

Reply: We corrected the typo.

Reviewer Point P 1.13 — p.5, l.88: sensitivity to NO2 -> it is a general discussion. I suggest replacing "NO2" by "the trace gas under investigation"

Reply: We changed the text as suggested.

> In this case, the AMF can be interpreted as the instrument sensitivity to the trace gas under investigation for measuring that specific VCD.

Reviewer Point P 1.14 — p.5, l.110: "computationally efficiency" -> computational

Reply: We corrected the mistake.

Reviewer Point P 1.15 — p.6, l.133: 577nm -> add space

Reply: We corrected the typo.

Reviewer Point P 1.16 — p.6, l.137: mostly 1000m resolution -> maybe more clear to describe it as a layer thickness instead of vertical resolution.

Reply: We changed the text as suggested.

> For the simulations 17 vertical layers were used with a thickness of 100 m below 1000 m and a thickness of mostly 1000 m above (see Table 1 in Wagner et al., 2007).

Reviewer Point P 1.17 — p.8, l.161: The upper row of Figures 3 (scenario at 577 nm) and 4 (scenario at 360 nm)

Reply: We changed the text as suggested.

The upper row of Figures 3 (scenario at 577 nm) and 4 (scenario at 360 nm) shows MYSTIC 1D-layer AMF profiles for the selected scenarios with a low elevation angle of 3° and a high elevation angle of 90° (zenith) without and with aerosols, respectively.

Reviewer Point P 1.18 — p.8, l.162: shows

Reply: We corrected the typo.

Reviewer Point P 1.19 — p.11, l.13: for -> at

Reply: We implemented the suggested change.

Reviewer Point P 1.20 — p.13, l.257: For clarity, mention explicitly GRAL is a dispersion model, eg. Graz Lagrangian dispersion model

Reply: We changed the text as suggested.

The NO_2 plume was computed with the Graz Lagrangian dispersion model (GRAL) (Oettl, 2015) for a 262.5 m tall stack located at x=1.9 km and y=1.3 km.

Reviewer Point P 1.21 — p.14, l.289: planet boundary layer (PBL) -> boundary layer has occurred many times earlier in the paper. Please explain PBL acronym at first occurrence and use the acronym in the continuation of the work.

Reply: We changed the manuscript accordingly.

Reviewer Point P 1.22 — p.15, l.294: SCCs->SCDs

Reply: We corrected the typo.

Reviewer Point P 1.23 — p.16, l.302: . . . 3 instrument zenith angles – > 3 viewing zenith angles (VZA)

Reply: We changed the text accordingly.

This is further illustrated in Fig. 9, where 2 of the 3 illustrated direct paths (i.e. 3 viewing zenith angles) cross the NO_2 maximum (main photon path (1) and (3) in Fig. 9)

Reviewer Point P 1.24 — p.16, l.310: (Fig 8 2nd row) ->(Fig. 8d, 8e, 8f) (same for line 312)

Reply: We changed the text accordingly.

Reviewer Point P 1.25 — p.18, l.339: The VCD cross sections

Reply: We corrected the typo.

Reviewer Point P 1.26 — p.21; l.374: the application of

Reply: We corrected the typo.

Reviewer Point P 1.27 — The spherical regression line and points are not clear at all. Maybe consider having two scatter plots. However, the main message of the plot stays clear based on the 5% deviation lines

Reply: We separated the one scatter plot in two scatter plots.

Reviewer Point P 1.28 — Figure 5 caption: Decay of vertically integrated AMFs with distance to the instrument (c) -> pease add"is visualized" after "instrument"

Reply: We modified the caption as suggested.

Reviewer Point P 1.29 — Figure 8: ...located at x=19 km and y=13 km. -> should be x=1.9 km and y = 1.3 km?

Reply: We corrected the typo.

Reviewer Point P 1.30 — Figure 12: maybe put "NO2 concentration"instead of "VCD" in plot and caption.

Reply: We modified the plot axis label to NO_2 column densities [$\mu mol/m^2$] and kept VCD in the caption because NO_2 concentration implies a point quantity. To be consistent with other figures, we changed the unit to $\mu mol/m^2$.

Reviewer Point P 1.31 — Table 2: Maybe better to give RAA instead of SAA in order to be consistent with the discussion at the end of p.19

Reply: RAA would not work in the table, because for pixels on the east of the instrument, have another viewing azimuth angle than pixels on the west for the same scene. We modified the text to be correct and consistent with the table.

Reviewer Point P 1.32 — Section 6 (Conclusion): I assume there is no need to define acronyms again here, e.g. AMF, RTM, VCD, SCD, etc.

Reply: We modified the text as suggested.

References

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Rozanov, V. and Rozanov, A.: Differential optical absorption spectroscopy (DOAS) and air mass factor concept for a multiply scattering vertically inhomogeneous medium: theoretical consideration, Atmospheric Measurement Techniques, 3, 751–780, 2010.

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