Impact of 3D radiative transfer on airborne NO2 imaging remote sensing over cities with buildings

Marc Schwaerzel et al. 2021

Response to the Reviewer's Comments

We thank Reviewer 3 for his/her positive comments, critical assessment and useful points to improve the quality of our paper. In the following we address his/her 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 — This paper describes the influence of 3D radiative transfer and shadowing on airborne NO2 retrievals, using a case study over Zurich. The paper is well-organized, concise and easy to follow. I have a couple of general and specific comments. After those are addressed I would recommend it be published in AMT. The study results are interesting but I am not sure how the results can be transferred over to a practical application in retrievals; however, it will be a good reference for the impacts of 3D radiative transfer on these kinds of measurements.

I find the description of the motivation to be unconvincing. The authors point to possible 3D effects as being responsible for the discrepancy between high resolution airborne NO2 maps over urban areas and city-scale urban models, and point to Figure S1 for an example. First, to drive home the need for this study, it would be good to see the motivating figures in the main paper instead of the supplement. To me, the observed and modeled NO2 maps look so very different that I doubt the source of the differences is entirely or even primarily 3D radiative effects. I would suspect issues with the model like inaccuracies in mixed layer height, transport, emissions, and chemistry, or issues with surface reflectance and profile shapes in the retrievals. For instance, there are what look like three plumes in the southwest corner of the map which actually look quite well-represented. Why are these represented fairly well but other plumes are not? It may be that 3D effects are the main reason for observed/model discrepancies, but the current example is unconvincing.

The study's motivation would be more convincing if: 1) The new calculations were actually applied to APEX data to calculate new AMFs, perhaps at the end of the paper, and the new maps showed better agreement with the model or improvements in resolution, or 2) a simulation were done using the GRAL NO2 columns to simulate airborne measurements that use 1D radiative transfer, and these simulations were found to show significant smearing. Even

i.e., along the lines of "we explore 3D effects as a possible contributor to smearing...". The later results will show whether or not they are significant.

Reply: We agree that difference between the APEX and GRAL NO_2 fields are not only caused 3D radiative transfer effects but also by limitations of the model, which is why it was not added to the main text. The cited conference presentation actually lists some likely necessary model improvements. We have removed the figure from the supplement and instead published the presentation containing the figure on Zenodo (https://doi.org/10.5281/zenodo.5220909). The suggestion to apply the method to APEX data and to compare the map to a GRAMM-GRAL simulation is currently work in progress and will be the topic of future publications. We have revised the introduction keeping a stronger focus on how 3D radiative transfer effects might contribute to spatial smearing and also added a subsection on real application implications:

A strong indication for the importance of 3D radiative transfer effects is that NO₂ maps obtained from airborne imaging spectrometers over cities are spatially much smoother than one would expect from the instrument resolution and compared to maps obtained from high-resolution city-scale dispersion models, which show, for example, strong gradients in the NO₂ field along major roads (Kuhlmann et al., 2017).

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Reviewer Point P 1.2 — My second general comment is that it would be nice to see some discussion of the practical implementation of these calculations. Is it too computationally intensive to use for actual campaign AMF calculations? How would a more realistic albedo field change the results?

Reply: We have added a new section in the paper showing the computation of VCDs from simulated SCDs and discussing the application to real observations as well as challenges (computational costs, realistic albedos and NO_2 fields, etc.):

The codes developed for this study can also be applied to real observations, for example, to the campaigns conducted with APEX imaging spectrometer. A major challenge is to obtain the required input data. 3D building data are available for many cities, but albedos for ground, roof and walls are generally not available. In addition, realistic 3D NO₂ fields from a building-resolving dispersion model are required to compute the total AMFs, which requires high-resolution emission inventories and additional model development, because most building-resolving models are not optimized for providing realistic vertical distributions of trace gases or cannot not be applied to a full city at high resolution (Berchet et al., 2017).

To minimize 3D effects when using 1D-layer AMFs, it would be recommendable to obtain the airborne spectrometer measurement around local noon when the SZA is lowest and avoid large viewing zenith angles. However, around noon turbulent atmospheric mixing will be strong and the NO₂ distributions would be smoothed as well.

The computation of 3D-box AMFs with buildings is computationally quite expensive, but still manageable for current airborne campaigns. For example, the computation of the 3D-box AMF field for a single

APEX pixel (e.g. on Fig. 6f) takes about 280 s on a single core of our Linux machine (Intel(R) Xeon(R) W-2175 CPU @ 2.50GHz). Processing a full campaign consisting of about 100'000 pixels takes about 23 days using all 14 cores on the system. However, simulating AMFs for an APEX campaign would not require simulation for a 1 km x 1 km domain. Nonetheless, computing 3D-box AMFs is significantly more expensive than computing 1D-layer AMFs and reducing computation time, for example, by finding suitable parametrizations using machine learning, would make it possible to calculate the 3D-box AMFs on smaller hardware, to larger campaigns or to run simulations with more details and at higher spatial resolution.

65 Reviewer Point P 1.3 — Line 3: It's unlikely that the entire cause would be 3D radiative transfer effects. Should qualify with wording like "3D radiative transfer effects may contribute to this discrepancy due to..."

Reply: Yes you are right that the wording could be misleading. We modified the sentence as following:

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This could partly be caused by 3D radiative transfer effects due to observation geometry, adjacency effects and effects of buildings.

70 **Reviewer Point P 1.4** — Line 152: Can you give an estimate (maybe in results section) about how much the results would change for shorter wavelength fitting windows? Shorter wavelengths are used in almost all other airborne and satellite instruments that measure NO2, so this would be most interesting for the majority of readers who do not use APEX data.

Reply: In general shorter wavelength increases scattering and decreases the instrument sensitivity. We simulated the footprint with a wavelength of 420 nm and without buildings to look at sensitivity distribution. It appears that 56% of the sensitivity is located outside the ground pixel. For simulations with buildings this number strongly depends on the buildings locations and the induced shadows and is therefore difficult to generalize. We added the following sentence in the conclusions:

In this study, the simulations were conducted at $490\,\mathrm{nm}$, which corresponds to the center of the fitting window used for NO_2 retrieval from the APEX airborne spectrometer. At shorter wavelength, used by other instruments, scattering increases, which decreases the instrument sensitivity to the main optical path. The footprint simulated with a wavelength of $420\,\mathrm{nm}$ (without buildings) shows the increase in scattering and the sensitivity to neighbouring pixels, as 56% of the sensitivity is located outside of the ground pixel.

85 **Reviewer Point P 1.5** — Some mixing of verb tenses in paper. For example, in abstract say "We compute.." then next sentence says "We found..."

Reply: We checked verb tenses in the manuscript and fixed the mistakes.

Reviewer Point P 1.6 — Line 63: I'm a bit confused at the wording of this sentence. Should it be "The model, however, is able to ..."

Reply: We modified the sentence as you suggested.

References

Berchet, A., Zink, K., Muller, C., Oettl, D., Brunner, J., Emmenegger, L., and Brunner, D.: A cost-effective method for simulating city-wide air flow and pollutant dispersion at building resolving scale, Atmospheric Environment, 158, 181–196, https://doi.org/https://doi.org/10.1016/j.atmosenv.2017.03.030, 2017.

Kuhlmann, G., Berchet, A., and Brunner, D.: High-resolution remote sensing and modelling of NO2 air pollution over the city of Zurich, in: 10th EARSeL SIG Imaging Spectroscopy Workshop 2017, Zurich, Switzerland, https://doi.org/10.5281/zenodo.5220909, 2017.