Answers to referee 1 comments "Impact of 3D Cloud Structures on the Atmospheric Trace Gas Products from UV-VIS Sounders – Part I: Synthetic dataset for validation of trace gas retrieval algorithms"

We thank the reviewer for the constructive comments that helped us to improve the manuscript. In the following we respond point-by-point to all comments.

This paper is one of a set of three interconnected papers that discusses a) a publicly available synthetic dataset of 3D radiances, b) the sensitivity of vertical column density NO2 retrieval errors near box-clouds and observations, and c) 3D cloud biases and metrics. The reviewed paper is part a) of the full set of papers. Since the paper is the first of a three-set collection, main findings are reserved for the other two papers. This produces the awkward situation that the main physical results, which one can derive from an analysis of the synthetic dataset, are not discussed in the reviewed paper. The reviewed paper is overly restrained. The reviewed paper is rather short of main findings, mainly stating that a synthetic data set is available, and therefore limited in informative results.

Yes, the main scientific findings are included in part II and part III. However, we think, that the description of how the dataset is created, which methods are used etc. is worth a publication. To our knowledge this is the first synthetic dataset including high resolution spectra (as used for trace gas remote sensing) and three-dimensional realistic clouds, generated using a three-dimensional radiative transfer model.

There are places in the text in which a terse one sentence paragraph is stated. Additional sentences can and should be added to the text in these portions of the text.

We have included several additional explanations, e.g. we explained in more detail, how the layer-AMFs can be used to study effects of the NO_2 profile on the retrieval error:
"Using MYSTIC, we may study how the layer-AMFs are modified by scattering from clouds in the neighborhood. Comparing the layer-AMFs of a clear sky atmosphere with the layer-AMFs influenced by clouds, we may estimate the retrieval error of, e.g., NO_2 vertical column densities (VCDs). Working with simulated layer-AMFs allows us also to study the impact of the vertical NO_2 concentration profile on the retrieval error. Since the influence of trace gases on the photon pathlength distribution and thus on layer-AMF is negligible, we may use the layer-AMFs of one radiative transfer simulation to estimate the error for various assumed NO_2 concentration profiles."

A suggested addition to the revised paper would be to include a figure or two that demonstrates the in-scattering and shadow curves (similar to Figure 2) for clouds of e.g. three heights in the LES cloud field, including panels in which (on the y axis) the reflectance is graphed as a function of distance from cloud edge, and panels in which retrieved NO2 is graphed (on the y axis) as a function of distance from cloud edge. These figures would help the research community better appreciate the quantitative

importance of 3D cloud effects upon NO2 retrievals. Over what km range are 3D effects present as a function of cloud height, and what are the column for these situations? I would have liked to have seen in the Conclusions section a discussion of the major physical findings of the paper. The Figures provide instructive insights, yet these insights are barely touched upon in the Conclusions.

A detailed analysis using the synthetic dataset based on LES clouds is presented in the third paper (Kylling et al., 2021), which includes several figures showing the impact of 3D cloud scattering on the NO_2 VCD retrieval error. Since the paper is available also in AMT, we do not like to duplicate these figures.
We have included a summary of the scientific findings from this analysis to the conclusions:
"The dataset based on LES clouds has been used to quantify the NO_2 VCD retrieval error. An operational retrieval algorithm was applied on the synthetic observations and the retrieval results were compared against the true NO_2 VCD which is the known model input. The exemplary results show underestimations of the retrieved NO_2 VCD in cloud shadow regions of more than 20% and overestimations of about the same order of magnitude in in-scattering regions for the specific sun-observer geometry. In the third paper of the series (Kylling et al., 2021), an analysis of the complete dataset is presented. Cloud shadow fraction, cloud top height, cloud optical thickness, NO_2 profile, solar zenith and viewing angle have been identified as the most important parameters determining the impact of cloud scattering on the NO_2 VCD retrieval. For low-earth and geostationary orbit geometries, 89 and 93%, respectively, of the retrieved NO_2 VCD were within 10% of the actual VCD for solar zenith angles less than 60°. For a solar zenith angle of 60° the numbers decrease to 53 and 61%. It was also found that for solar zenith angles less than 10°, the NO_2 VCD retrieval error is generally smaller than 10%. For larger solar zenith angles the retrieval error increases to values of the order of tens of percent."

Page 3, line 12. Clarify what is meant by "bias" (the bias of what?)
Page 3, line 12. Replace by "In the third paper by Kylling..". The one sentence paragraph is a bit jarring since it is overly short in informative content.

Rephrased and merged 2 paragraphs summarizing the third paper of the series Kylling et al. (2021).

"In the third paper by Kylling et al. (2021) the NO_2 VCD retrieval error due to 3D cloud scattering has been quantified using both, synthetic and observational data."

Page 4, line 3. Clarify what is meant by "unbiased radiances"

Removed "unbiased" because it is not clear from the context.

There are variance reduction methods, e.g., the so-call phase function truncation method, which cause a bias in the computed radiances. The VROOM methods are physically correct, do not use approximations and therefore the results are not biased.

Page 4, line 6. Replace "agreed perfectly" with a quantitative value.

We provide six references to different model intercomparison studies. Depending on the specific study and on the investigated quantities, the level of agreement is not always the same, so we can not provide one quantitative value here, the reader needs to look into the given references. Removed the word "perfectly", because this might be a misleading term.

Page 4, lines 14-17. I am not convinced of the ability of the authors to "calculate the full spectrum based on photon path distributions sampled at a single wavelength". Atmospheric optical properties (Rayleigh scattering, aerosol optical depths, asymmetry and single scattering albedo) have a wavelength dependence in a real atmosphere. Please support your statements in the context of a real atmosphere with additional sentences. The sentence "The statistical error of such a simulation is a bias for the complete spectrum" is not comprehensible. Add additional sentences which discuss the ALIS method. Replace with "This method allows one to calculate .."

Included some additional sentences to describe the ALIS method. Please refer to Emde et al. (2011) to understand the details, this paper includes also a validation of the method.

"The Absorption Lines Importance Sampling (ALIS) method (Emde et al., 2011) solves this problem. This method allows one to calculate the full spectrum based on photon path distributions sampled at a single wavelength. In order to take into account the spectral dependence of the absorption coefficient a spectral absorption weight is calculated for each photon path. Further, at each scattering event the local estimate method (Marshak and Davis, 2005) is combined with an importance sampling method to take into account the spectral dependence of the scattering coefficient. Since each wavelength grid point is computed using the same photon path distribution, the statistical error of such a simulation is is almost independent of wavelength, i.e. it corresponds to a small offset of the complete spectrum. For DOAS type retrievals this error is completely removed by the polynomial fit to compute the differential optical thickness. This statistical error decreases with the number of photons used in the simulation and converges towards the correct spectrum. The method it is very well suited to efficiently simulate radiance spectra in high-spectral resolution."

Page 5, line 1. What is the boundary layer height, and how is NO2 vertically distributed in the troposphere?

Included a more detailed description of the model atmosphere (including vertical layering) and also a figure showing the NO2 profile.

Page 5, line 10. Change to "in the x-direction".

Done.

Page 5, line 14. The sentence implies just a single box-cloud geometry, while Table 1 lists several box-cloud heights. Rephrase to "For the liquid water cloud the primary cloud geometry has the base height set to 2 km ...".

This sentence refers to the base case, as stated in the sentence before.

Clarified later on that we start with the base case and vary the different parameters:

"Starting from the base case, we varied the following parameters: cloud optical thickness, cloud bottom height, cloud geometrical thickness, solar zenith angle, and surface albedo."

Page 6, line 4. Why were aerosols not included in the calculations?

We included the following explanation:

"Aerosols were not included, although aerosol scattering also has a significant impact on the NO_2 retrieval. However, in this study, we aim to quantify the impact on cloud scattering on the retrieval. When both, aerosols and clouds are included, it becomes difficult to disentangle the impacts of cloud and aerosol scattering. Therefore, we decided to include only clouds."

Page 6, line 11. Clarify what is meant by "variance reduction methods". Add sentences that describe the VROOM methods.

Included a short explanation:

"The variance reduction methods VROOM (Buras and Mayer, 2011), which reduce the statistical noise in Monte Carlo radiative transfer simulations including cloud scattering,"

Detailed explanations are out of scope of this paper, the reader is refered to Buras and Mayer (2011), which describes all methods thouroughly.

Page 9, line 12. Change to "Note that D(l) is a smooth function"

Done.

Page 11, line 8. Change to "layer-AMF as a function of"

Done.

Page 13, line 2. Change to "realistic 3D clouds"

Done.

Page 15, line 17. Replace "sufficiently accurate" with a quantitative accuracy.

Included "reflectances agree to 3 digits after the decimal point ".

Page 15, line 22. Change to "was analyzed, and it was found that SZA.."

Done.

Page 16, line 1. Change to "It was found that SZA varies.."

Done

Page 17, line 1. Change to "Note that each simulated pixel includes 36 cloud pixels,.."

Done.

Page 17, line 5. Change to "Note that the number of .."

Done.

Page 18, Figure 11. What are the units of the NO2 retrieval error?

The figure shows the relative error (no unit).

Page 19, line 16. Change to "Note that the complete LES.."

Done.

Page 23, line 7. Explain how the synthetic dataset can be used to "validate the various different trace gas retrieval approaches for Sentinel-S5P." This is an example of a terse one sentence paragraph that would benefit from additional sentences.

Included additional sentences: "The algorithms can be applied on the synthetic data. Comparing the retrieved NO_2 VCDs to the true value used as input to the radiative transfer simulations yields the retrieval accuracy of each algorithm."

References

Buras, R. and Mayer, B.: Efficient unbiased variance reduction techniques for Monte Carlo simulations of radiative transfer in cloudy atmospheres: The solution, J. Quant. Spectrosc. Radiat. Transfer, 112, 434–447, 2011.

Emde, C., Buras, R., and Mayer, B.: ALIS: An efficient method to compute high spectral resolution polarized solar radiances using the Monte Carlo approach, J. Quant. Spectrosc. Radiat. Transfer, 112, 1622–1631, 2011.

Kylling, A., Emde, C., Yu, H., van Roozendael, M., Stebel, K., Veihelmann, B., and Mayer, B.: Impact of 3D Cloud Structures on the Atmospheric Trace Gas Products from UV-VIS Sounders – Part III: bias estimate using synthetic and observational data, Atmos. Meas. Tech. Discuss., submitted, 2021.

Marshak, A. and Davis, A.: 3D Radiative Transfer in Cloudy Atmospheres, Springer, iSBN-13 978-3-540-23958-1, 2005.