Answers to referee 2 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 constructive comments that helped us to improve our manuscript.

In the following we respond point-by-point to all reviewer comments.

The manuscript reads often like a technical report, sometimes difficult to follow. The fact that it presents a synthetic dataset as a part of three publications decreases the possibility of more in depth and detailed discussions. However, scientific discussion needs to be more extensive, otherwise it could just be published in a journal more suitable for datasets.

The main scientific findings are included in part II and part III, this is correcrt. 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.

Two main issues that are of central importance (together with clouds) to trace gas retrievals are aerosols and NO2 profile. The aerosols and its correction on the retrieval algorithms is closely related to the clouds and their effects. Furthermore, the study uses the most realistic cloud field possible with 3D radiative transfer model but then uses a constant NO2 field throughout the domain. These two topics need to be discussed in the text, as currently is just mentioned as if this was of very little relevance.

See answers below.

Introduction: The first two-three paragraphs as compared to the rest of the introduction are poorly written. The last sentence of the first paragraph does not add anything to the readability of the introduction and to the topic of the manuscript.

Removed the last sentence from the first paragraph. Slightly changed the other two sentences.

Page 2, lines 24-26: "Here" referring to Schwaerzel et al? What do you mean by 3D box-airmass-factors? Calculated with 3D radiative transfer model? Please be more specific. This is the introduction and it is already somewhat confusing the naming of the AMFs. Later on, you also refer to 3D layer-AMFs (e.g. page 19, line 20), so please be consistent.

Removed this statement from the introduction and included more detailed definitions of layer-AMF and box-AMF in the model description section. Here we also shorty explain the DOAS method, how the layer-AMFs are used in the retrieval, and how layer-AMFs can be used to study the impact of cloud scattering on the retrieval.

"In the UV and visible spectral ranges, the standard retrieval algorithm is based on the DOAS technique Platt (2017): in a first step, the slant column density (SCD) is retrieved by spectral fitting of the observed solar spectra to absorption cross sections of trace gases. The SCD corresponds to the amount of trace gas along the average photon path from the Sun through the atmosphere to the satellite sensor. In order to convert SCD into a vertical column density (VCD), the so-called air-mass factor is required, which is defined as the ratio between SCD and VCD. In clean regions, the retrieval error is dominated by the spectral fitting, while for polluted or cloudy regions, the uncertainty of the AMF becomes the dominant error source. The AMF is calculated using radiative transfer models.

MYSTIC includes the option to simulate 1D layer-AMFs or 3D box-AMFs (Schwaerzel et al., 2020). The concept of layer/box-AMFs assumes that the trace gas concentration is small compared to the concentration of other gases, meaning that interaction of photons with trace gas molecules does not alter the photon path distribution in the atmosphere. Layer-AMFs are calculated from the photon path length distribution in each individual altitude layer of the model atmosphere as described in Deutschmann et al. (2011). MYSTIC allows to calculate layer-AMFs for 1D plane-parallel or spherical atmospheres, and also for 3D model atmospheres. In the latter case the photon pathlengths are integrated horizontally over the full domain. Note that these "3D" layer-AMFs still include the impact of 3D cloud scattering. In DOAS type retrievals the layer-AMFs are used together with the a priori NO_2 altitude profile to compute the total AMF:

$$\mathrm{AMF} = \frac{\sum_l \mathrm{AMF}_l \cdot x_l}{\sum_l x_l} \tag{1}$$

Here l is the layer index, AMF_l the layer-AMF and x_l the partial column density for layer l. This AMF is then used to convert from slant column density (SCD) to vertical column density (VCD):

$$\mathrm{VCD} = \mathrm{SCD}/\mathrm{AMF} \tag{2}$$

Note that in the literature, layer-AMFs are commonly called box-AMF (e.g. Deutschmann et al. (2011)), which is a confusing terminology, because they do not refer to model grid boxes. MYSTIC also enables the calculation of real "box"-AMFs which are derived from the 3D photon pathlength distribution, i.e. from the photon pathlengths in each 3D model grid cell. Box-AMFs are useful if one knows a 3D a priori NO_2 concentration distribution which can be used in the retrieval to convert from SCD to VCD (Schwaerzel et al., 2020). All currently available operational retrieval algorithms apply 1D a priori altitude concentration profiles, therefore they can not use box-AMFs.

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. Such an analysis is presented in part II of this publication series (Yu et al., 2021). For this reason it is not necessary to include simulations for different NO_2 profiles in the synthetic dataset."

Page 3, line 33: what do you mean by inhomogeneous surface albedo? Spectral dependence? Reflection anisotropy?

We actually meant a 2D surface albedo map. But spectral dependence and reflection anisotropy can also be handled. Included a more precise description in the text.

Page 4, line 15: is the ALIS method influenced by the number of photons on the simulation?

Yes, the bias decreases with the number of photons. This statement has been added to the text.

Page 4, line 18-19: see my comment on the introduction about layer and box AMFs.

See answer above.

Last sentence on Sect. 2: maybe you can cite the paper of the series where this is actually analyzed. In this manuscript only one specific layer AMF has been analyzed (at 0.5 km).

Yes, a more detailed analysis is presented in part II of the series (Yu et al., 2021). The reference has been added to the text.

Sect. 3: Page , line 1: 'most of the NO2 located within the BL'. How high is the boundary layer for your base case? Have you investigated the effects in a non-polluted atmosphere?

We included a figure showing typical NO2 profiles, including the polluted one which was used for the base case. We also investigated effects for a non-polluted atmosphere (actually here the effects are much smaller). This can be done using the layer-AMFs, which do not depend on the NO2 profile, as long as NO2 molecules do not change the photon pathlength distribution. Generally this is a good assumption for trace gases.

We included a justification for including only one NO2-profile in the synthetic dataset: "As mentioned before we may use layer-AMFs to investigate the impact of cloud scattering on the trace gas concentration retrieval. Layer-AMFs are independent of the trace gas profiles, for this reason we define only one NO_2-profile, but still we can investigate retrieval errors also for different profiles including non-polluted cases (see also Yu et al. (2021))."

In connection to the apriori profile and the horizontal effects; the TROPOMI NO2 bias as reported by validation studies is different for urban and rural areas, so the 3D clouds effect may play a different role in these biases depending on the pollution level. See also your sentence in page 12, line 11. This needs further discussion, 'more or less affected' is not rigorous.

Yes, of course the retrieval error depends a lot on the NO2 profile. We investigated this in detail using the synthetic data. At the end of section 3 we include a brief summary of the sensitivity study based on the synthetic data for the box cloud case which is presented in detail in Yu et al. (part II).

"A detailed sensitivity study of the NO_2 retrieval error based on the box-cloud synthetic dataset is presented in Yu et al. (2021). Largest retrieval biases were found in the cloud shadow region, typically the errors are in the range of 10–100% for the polluted scenario. The bias increases with solar zenith angle, decreases with surface albedo and it increases with cloud optical thickness. The dependency on cloud geometrical thickness and cloud bottom height is less pronounced. Yu et al. (2021) also show that the cloud effects are much stronger for polluted cases compared to non-polluted cases, the maximum retrieval bias for the polluted profile is 95% for the base case settings and for the clean profile it is reduced to 6%. Various different NO_2 profile shapes have been investigated in addition, clearly demonstrating that the retrieval bias depends on the altitude where most of the NO_2 is located. The synthetic data was also applied to investigate the dependancy of the retrieval bias on the spatial resolution of the instrument. The synthetic data is created for a sensor footprint of $1\times1\,\mathrm{km}^2$. By averaging, spatial resolutions between 3-15 km could be investigated. As expected, the retrieval bias decreases with increasing spatial resolution due to spatial averaging. The cloud shadow effect strongly depends on the cloud shadow fraction in a pixel."

Later on the section it is also mentioned that "Aerosols are not included". Aerosols are a relevant topic for NO2 retrievals, so it needs further discussion. Even if it is not included, some motivation for this decision should be discussed, as well as the effect that the inclusion of an explicit aerosol treatment would have in the results.

Included the following discussion:
"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."

What is the vertical discretization of the atmosphere? How does affect your simulations?

Included the following sentence for clarification:

"We have chosen a fine vertical resoltution of the model atmosphere in the lower part of the atmosphere, between 0 km and 12 km altitude the layer thickness is about 150 m. The vertical resolution from 12–25 km is 1 km, from 25–50 km 2.5 km and from 50–100 km 5 km. We have chosen the fine vertical resolution in the lower part of the atmosphere in order to resolve the vertical dependency of layer-AMF in the region of interest."

Sect. 3.1.2: what do you mean that in the y-direction the cloud layer extended to infinity? As I later understand the cloud has a geometrical dimension, right? If half of the domain extending from 0 to 100km is cloud-free and the other half has a cloud, this does not match the schematic in Fig. 1.

MYSTIC uses periodic boundary conditions, this information has been added to the text. It means that next to the cloud, there is again a clear region, so the schematic was in principle correct. However, in order to clarify the setup, we updated the schematic so that half of the domain is clear and half cloudy, and we show the in-scattering and the shadowing geometry separately, using the same cloud definition but changing the sun direction exactly as done in the simulations. The y-direction really extends to infinity as stated in the text.

Page 6, line 3: above the cloud as in the vertical dimension?

Changed this formulation to "... starting at a distance of 15 km away from the cloud egde in the clear region and ending at a distance of 10 km in the cloudy region."

Page 7, line 7: "the reflectance is higher than the clear sky reflectance near the cloud edge." Reads weird, please rephrase.

Rephrased to: "In the in-scattering region (left panels), the cloudy reflectance is larger than the clear sky reflectance."

Page 6, line 7: 1D cloud layer setup. This is the first time this is mentioned. Do you mean just a cloud acting as a Lambertian reflector? Please explain.

No, we use exactly the same cloud definition (same optical properties, liquid water content etc.) but extend the cloud layer over the full domain. Clarified this in the text.

"For all combinations of parameters we also calculated radiance spectra for a corresponding 1D cloud layer setup, where the cloud is extended horizontally over the full model domain. The cloud optical and microphysical properties are exactly the same as for the 3D cloud simulations."

Page 6, line 11: what are variance reduction methods, why are they needed? Please explain.

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," Explaining the methods in detail is out of the scope of this paper, the reader needs to refer to the given reference.

Reading of figure 1 could benefit from the addition of a grid. On page 9, line 31: do you mean larger than -15%?

We added the x and y axes to figure 1 for clarification. Yes, we mean -15% and corrected this.

Sect. 3.2.3. The explanation on this section could benefit from an Eq. that shows how the AMF is used in the retrieval.

We included in the model description part along with the definition of the layer-AMFs the basic equations used in the retrieval to convert from SCD to VCD.

Last sentence in page 12: it would be beneficial to include a discussion with few sentences on the main findings even though they are published in Yu et al.

As suggested a short summary of findings presented in Yu et al. 2021 has been included.

Page 15 line 29: TROPOMI was not launch until October 2017.

The year is actually not used to calculate the sun-observation geomtry, it is the same every year. Omitted "2017" in the text.

Sect. 4.2.3: I would suggest to substitute the global maps of surface albedo for a zoom over the study for which the cloud simulations are done.

Done.

Sect. 4.2.4: What about higher resolution than 7 km x 7 km? This is good for TROPOMI, but future sensors will definitely provide measurements at higher resolution. In Sect. 3 the simulations are done for higher resolution, and the ICON clouds resolution is 1.2 km x 1.2 km. The increased spatial resolution (as pointed in the manuscript) will enhance the impact of 3D clouds effects, so it would benefit the discussion to perform these simulations at higher resolution. If this is not feasible, then at least this should be discussed.

Higher spatial resolution would indeed be interesting. However, the aim of the 3DCATS study was to quantify the retrieval error for TROPOMI, therefore we generated the synthetic data for TROPOMI and not for future instruments. The impact of spatial resolution has been investigated using the box cloud synthetic data. This is discussed in Yu et al (part II). A reference to this has been added to the end of Section 3 of this paper.

What is the effect on the results of reducing the number of photons with respect to the 1D/3D case? How will this affect the airmass factor calculation?

The standard deviation of the Monte Carlo results increases, clarified this in the text:

"Note, that the number of photons was 100 times less than for the box-cloud and clear-sky cases presented in Section 3. Therefore, since the standard deviation of a Monte Carlo simulation is inversely proportional to the square root of the number of photons, the standard deviation of all simulations results (reflectance spectra and layer-AMFs) is increased by a factor of 10."

The study uses a very realistic cloud field from LES simulations, but then assumes a constant NO2 field over the whole domain, which is very unrealistic. The consequences of this assumption on NO2 needs to be discussed. How would the NO2 retrieval error on Fig. 12 look like if a realistic NO2 field would be used?

The retrieval error in Fig. 12 would look completely different if we use an inhomogeneous NO2 field, errors would be small in clean regions and larger in polluted regions. It would be very difficult to relate the results to the cloud properties. For this reason we decided to include a homogeneous NO2 profile in the background atmosphere. Anyway, we also provide the layer-AMFs which can be used to investigate different NO2 profiles.

We included the following statement as explanation: " We have chosen a constant NO_2 profile because we aim to investigate the impact of realistic clouds on the retrieval results. When we include an inhomogeneous NO_2 profile it is not easily possible to quantify this impact, e.g. to figure out, which type of clouds have the largest impact on the retrieval error. This is only possible when we have the same atmospheric background conditions over the full domain."

Figures and figure captions should be revised. Different sub-figures are specified differently in different figures, so please revise. See https://www.atmospheric-measurement-techniques.net/submission.html for figure guidelines. Using letters

a,b,c etc. makes referencing on the text easier. Please mind the reader when creating the figures. For example, Fig. 10: '(similar to TROPOMI on Sentinel-5P and Sentinel-5)' this is not relevant in a figure caption. Lower panels 'x = 256 km' is not relevant information and makes the figure busier. Another example, Fig. 3 Top: legend 'clear' is better understood than '-1.5' and '-10.5 km', maybe add 'shadow' and 'clear region'.

We revised the figures and included letters to refer to the panels in the figures. Further we revised several legends and figure titles as suggested.

Name the O2 A band consistently throughout the manuscript (three names O2-A band, O2A-band, O2A band have been used)

Named "O_2A-band" consistently.

Page 2, line 28: for->from TROPOMI/S5P obs.

Done.

Page 2, line 29: synthetic -> synthetic

Done.

Page 2, line 30: are-> were not included

Done.

Page3, line 3: incorrect grammar; the bias due to 3D clouds on what? And no need to start new paragraph if you talk about the same paper. Rephrased.

Page 18, limes 5-10: please write sentences in present tense. E.g., "pathlength is decreased" -> "decreases"

Done.

Short paragraphs (1-2 sentences) just expressing technical details should be avoided.

Merged the short paragraphs.

Please include (at least) a reference when mentioning FRESCO cloud algorithm.

Done.

Page 22, line 6: what about the shadow effects?

Rephrased: "... including effects of clouds (in-scattering and shadowing) ..."

Page 22, line 23: was this not at 1.2 km x 1.2 km?

Yes, thank you. Corrected this.

References

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