

We gratefully thank the reviewer for the careful reading of our revised manuscript and for the constructive comments. Below the reviewer's comments are reproduced in italic bold font. Our responses to these comments are given in roman font and pieces of text added to the manuscript are displayed in blue font.

Referee #1:

The authors have addressed many of the points raised and have added relevant additional evaluation for clouds as layers which is nice.

Two of my comments appear to have been misunderstood:

"It would be good to have a short discussion of what the expected effect of aerosols is on the discussed 3d effects which here are discussed in a Rayleigh atmosphere."

The authors reacted by briefly discussing the relevance of aerosols in NO₂ retrievals, but my intention was to reflect in how far the assumption of a Rayleigh atmosphere may have an impact on their results. For example, scattering aerosols in a cloud shadow may have different impacts on NO₂ AMF than scattering aerosols outside the cloud shadow. Maybe the authors can give their thoughts on how relevant the neglect of aerosols is for their results.

This discussion on the impact of aerosols has been removed from the introduction and rewritten under section '2.2 cloud correction':

"Aerosols are not included in this study. However, the presence of aerosol may lead to different impacts on the 3D effects, depending on aerosol properties, such as single scattering albedo, optical depth and vertical distribution. For example, scattering aerosols in the cloud shadow will increase the AMF and compensate the shadowing effect, whereas strong absorbing aerosols may decrease the AMF and enhance the 3D effect. The resulting effect may be rather complex, and further investigation would be needed for an accurate evaluation of such effects. In addition, it should be noted that, in practice, aerosols are implicitly treated as clouds in actual retrievals since the effects of aerosols are expected to be similar to those of clouds (Boersma et al., 2004, 2011)."

"The application to TROPOMI data is based on the assumption that NO₂ retrievals should yield the same column in cloudy and clear regions as well as in the cloud shadow. However, considering the reduced actinic flux in the cloud shadow (and the increased values inside the cloud), shouldn't we actually see differences?"

The authors react with a reference to 3d-AMFs but my point here is, that it is not trivial to assign low NO₂ in the cloud shadow to a retrieval problem as less NO₂ is expected in non-illuminated regions. I think this fact should be mentioned in the discussion in section 4.3

It is correct that we cannot rule out that photochemical effects take place in the cloud shadow/cloudy regions and locally modify the NO₂ concentration. However, a reduction in actinic flux would probably lead to an increase of the NO₂ concentration due to a shift in the NO-to-NO₂ ratio. In principle, the independent pixel approximation approach in the AMF calculation may account for this by using intensity weighted AMFs from cloudy, clear and cloud shadow parts. In reality it is however not possible to treat such effects in a rigorous way, since the overall impact of a change in illumination on the NO_x chemistry cannot be quantified in a simple way. Spatial variability (due to chemistry or transport) is certainly important and responsible for significant uncertainties in the retrieval. In this study, TROPOMI observations were selected so that they can be divided into complete cloudy, clear and cloud shadow conditions. The uncertainty of the NO₂ retrieval is discussed in the last paragraph.

In order to better discuss the importance of spatial variability, I added to following statement in section '3.4: Change of spatial resolution':

“Note that the synthetic data used in this study assumes that the NO₂ column is the same in clear and cloudy regions as well as in cloud shadow. Consequently, the NO₂ retrieval is based on the same assumption. In reality, however, the NO₂ column usually shows significant to large horizontal variability, which leads to uncertainty in the retrieval. The importance of such effects cannot be easily assessed using tools available for this study, and would need to be further investigated.”

I also note that there are a few typos in the newly added text and that some figure references now are no longer correct.

Done

Referee #2:

I still see an inconsistency between the sophisticated investigation of 3d effects on NO₂ AMFs on the one hand, and at the same time treating cloud effects following a simple independent pixel approximation.

This study aims to investigate the impact of 3D cloud structures on the trace gas retrieval products from UV-Visible sensors. The current operational trace gas retrievals are based on the DOAS approach, which consists two steps: DOAS spectral fitting methods to get SCD, and conversion of SCD into VCD by means of calculated AMFs. The 3D effects mainly affect the calculation of the AMF, and the cloud correction in AMF is based on the independent pixel approximation. This approach could not capture all 3D effects, and this study is to identify which situation will produce significant bias due to 3D effects, and investigate possible (simple) mitigation strategies for such cases.

I don't understand at all how the authors can state that "The accuracy of cloud retrieval does not link to the accuracy of cloud correction" (reply to Referee #2), as clouds are strongly affecting the AMF, so their accuracy definitely matters.

“Examples of cloud and NO₂ retrieval are shown in Figure A1. The O₂-O₂ and FRESCO cloud fraction retrievals show very good agreement. However, cloud pressure retrievals show large differences, especially for high cloud cases. It should be noted that the cloud pressure retrievals based on O₂-O₂ or O₂ absorption must be interpreted effective values. The accuracy of the cloud retrieval does not always link to the accuracy of the cloud correction in the NO₂ retrieval. For instance, the O₂-O₂ cloud pressure substantially differ from true values for the high cloud cases, whereas FRESCO cloud pressures are usually compared to the middle of the cloud layer. On the other hand, NO₂ AMF using an O₂-O₂ correction are often closer to the true AMF than those using FRESCO correction.”

Rephrased by:

“Examples of cloud and NO₂ retrieval are shown in Figure A1. The O₂-O₂ and FRESCO cloud fraction retrievals show very good agreement. However, cloud pressure retrievals show large differences, especially for high cloud cases. It should be noted that cloud pressure retrievals based on O₂-O₂ or O₂ absorption must be interpreted as effective values. Furthermore, a more accurate cloud retrieval does not always correspond to a better cloud correction in the NO₂ retrieval. For instance, the O₂-O₂ cloud pressure substantially differs from true values for the high cloud cases, whereas FRESCO cloud pressures are usually compared to the middle of the cloud layer. On the other hand, NO₂ AMFs using an O₂-O₂ correction are often closer to the true AMF than those using a FRESCO correction. These results also show different impact on the retrieval between the polluted and clean cases. It implies that the accuracy of the cloud correction relies not only on the accuracy of the cloud retrieval, but also on other factors, such as the NO₂ profile.”

I also wonder if the authors have understood this fundamental problem, as in their reply, they refer to additional figures that show solely 1d cloud retrieval results. I don't understand how this 1d analysis makes the authors "believe that the analysis of 3D effects on cloud products is not a relevant topic in this paper".

So I still see a problem in the study design: 3d effects are solely analysed with respect to trace gas AMFs, but not on the cloud retrievals.

Thus the authors should clearly state this inconsistency in their study in abstract and introduction, and should discuss how far this inconsistency could be resolved in future studies in the discussion/conclusion.

The aim of this study is to assess the performances/limitation of current trace gas retrievals. Therefore, cloud correction approaches used in the retrieval are based on simplified cloud correction schemes for all cloud effects, including 3D effect.

1D cloud/NO₂ retrievals show that the accuracy of the cloud corrections may depend on many factors, and it's therefore difficult to assess the accuracy of the cloud correction only based on an analysis of the accuracy of the cloud retrievals.

In addition, some descriptions of cloud product used for NO₂ retrieval and cloud retrievals in cloud shadow/in-scattering regions are added:

Introduction (Line 86): "The present paper focuses on impact of 3D effects on the classic tropospheric trace gas retrievals, including identification and investigation of the significant retrieval biases due to the 3D clouds, and exploration of mitigation strategies for these cases."

Methodologies (Line 126): "Notice that, all cloud effects, including the 3D effect, are treated based on such simplified cloud correction schemes, however, these approaches may not capture all cloud effects, which leads to uncertainty in the NO₂ retrieval."

NO₂ retrieval in the vicinity of a box-cloud (Line 268): "For these pixels, the retrieved CFr is greater than 0 due to the enhanced reflectance, and the O₂-O₂ value is slightly higher than that of FRESCO. Cloud pressure retrieval is usually a bit lower than surface pressure, but higher than neighboring cloud pressure, and the FRESCO cloud pressure is relatively higher (not shown)."

Conclusions and Outlook (Line 566): "The cloud products used in the NO₂ retrieval treat the cloud shadow pixels as cloud-free, resulting in large positive biases (up to more than 100%) in the NO₂ AMF calculation."

Future work includes (Line 585): "Another mitigation method is to develop more sophisticated cloud retrievals, which account for the 3D effects, are feasible to apply to satellite observation, and can easily adapt to trace gas retrieval algorithms"

Bibliography

Boersma, K. F., Eskes, H. J., & Brinksma, E. J. (2004). Error analysis for tropospheric NO₂ retrieval from space. *Journal of Geophysical Research: Atmospheres*, 109(4). <https://doi.org/10.1029/2003jd003962>

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