

Interactive comment on “On the improvement of NO₂ satellite retrievals – aerosol impact on the airmass factors” by J. Leitão et al.

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First of all we would like to thank the reviewer Folkert Boersma for his helpful review with very positive and beneficial comments that have certainly contributed to the improvement of this paper. We have integrated all the requests where possible and will provide necessary answers and explanations in the following text.

Comment: I see two limitations in this study: 1) the results hold for one class of retrievals, i.e. clear-sky situations without clouds. Since 90% (Krijger et al. [2007], and even more for instruments like GOME(-2) and SCIAMACHY) of satellite viewing scenes is partly or completely cloud-covered, this is a major limitation on the applicability of the results (and this limitation is not yet acknowledged in the manuscript), 2) even in sit-

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uations of clear-sky retrievals, it is still very difficult to generalize the results because the observed aerosol information used by the authors is only holds for a few particular places, and falls short of the aerosol fields needed for all local situations.

Reply: We agree with the reviewer that clouds are a major factor in the retrieval of tropospheric NO₂ columns. Depending on cloud fraction, cloud top height, cloud optical thickness and the internal structure of the cloud they can have diverse effects on the sensitivity of the measurements, and often are more important than aerosol effects. However, we feel that it is important to understand the effect of aerosols in the simplest case (no clouds) first before attempting to evaluate the more complex situations where clouds are also present. In the latter case, the details of the treatment of clouds in the retrieval will play a large role, and such a study is out of the scope of the present manuscript. Nevertheless, in the revised paper, we have added a qualitative discussion on cloud effects and have emphasized the importance of clouds for the retrieval.

The second point of the reviewer is the difficulty of generalization. The reviewer is right that this manuscript does not describe an algorithm to correct for aerosol effects in the retrieval, but rather aims at probing the sensitivity of the measurements to different aerosol scenarios. In this context, we have tried to collect a set of scenarios from real atmospheric measurements that are representative of as many different situations as possible. The limitations of this data set are mainly due to the lack of measurement data which could be used. We therefore think that we have already selected as good a data set as possible and do not know how to improve on it.

General points

Comment: The only general issue I see, is that the text repeatedly suggests that NO₂ retrievals are overestimated or underestimated ‘if the presence of aerosols is not taken into account’. This is true for clear-sky retrievals in the particular retrieval setting chosen by the authors, that is, without a cloud-correction scheme. To give the text of the manuscript a wider appeal, also addressing retrievals that explicitly correct for en-

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hanced light paths and top-of-atmosphere radiance, I suggest the authors rephrase the text in such a way to reflect that it is not the presence of aerosols per se, but rather the effect of aerosols on light paths and TOA radiance, that needs to be taken into account to avoid retrieval biases.

Reply: As mentioned above, we agree that clouds need more attention in the manuscript and have added a qualitative discussion on their impact and the role of correction algorithms. However, a detailed and quantitative treatment would be a study on its own and should be the topic of a separate paper. With respect to the rephrasing suggested by the reviewer we have tried to improve on the text in the basic description of the effects. Within the main text, we still use the formulation "effect of aerosols" as the more precise description would make the text difficult to read in places.

Comment: The paper would also benefit if the authors provide some discussion of a possible strategy to start generalizing their results; using satellite aerosol observations, or model information to overcome the limited spatial representativity of this study?

Reply: Certainly this is an important issue that we failed to address properly. We had only shortly mentioned in the conclusions and have now extended this part of the text. This is an important part of the study and a challenge for future work. However the different areas and topics to be explored in this subject are numerous. This could lead to a rather extensive conclusion compromising in part the conciseness but also the focus of the present paper and we have, therefore, limited the discussion to this paragraph.

Comment: also recommend investigating partly-cloudy scenes to extend beyond the clear sky only scenes?

Reply: As said above, we agree that this is an important and interesting topic but think that a quantitative treatment of all the complexities introduced by the various possible combinations of clouds and aerosols is out of the scope of this manuscript. However, we added a section with a qualitative discussion.

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Comment: The manuscript could be shortened; the first 15 lines of section 2.3 could be skipped. It should be sufficient to refer to the use of AERONET data here, without mentioning all the other networks. On page 3234, lines 13-28 could be shortened too, a reader gets the point that elevated aerosol layers are a fact-of-life, and there is no need to provide a complete literature overview here. P3235-3237, section 3 could be shortened too.

Reply: As data from other networks was also used for this study they are still mentioned in the text. Still, we agree with the referee that the manuscript gains by leaving some much detailed information out and focusing on the important details of the study. Based on this suggestion together with those from the second referee we have shortened the mentioned sections and improved the text.

Comment: In my opinion the reader could do without an outline of the results first, before actually getting to them.

Reply: Thank you for bring this point to our attention. We have followed the advice and removed this part from the original manuscript.

Comment: Table 1 is useful, but I found it inconvenient to have to page back and forth when interpreting the Figures showing the results. My suggestion is to include in the Figure captions the specific scenario used and what this means in terms of vertical profile, AOT, single scattering albedo, etc.

Reply: The figure captions where this information was missing include now further information on AOD and SSA per scenario.

Detailed points

Comment: P3226, l18: because scattering is first and foremost a physical process, I suggest to rephrase that 'for an ensemble of photons, scattering is regarded as a statistical process'.

Reply: The sentence was changed according to the suggestion.

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Comment: P3226, I22: typo, there should not be a comma after 'both'.

Reply: Corrected.

Comment: P3226, I22-23: this sentence is difficult to interpret. I guess the authors mean that 'relative to a pure Rayleigh atmosphere, aerosols change both the individual light path lengths and total radiance observed at the satellite'.

Reply: Corrected.

Comment: P3227, I18-20: the authors later show that viewing geometry can not be discarded, and I suggest to include viewing geometry in this listing.

Reply: We are not sure what the reviewer refers to with "viewing geometry". The SZA was missing and has been added to the list. However, the dependence on the instrument viewing direction was not studied here although the results will clearly be different for non-nadir viewing directions.

Comment: P3229, I15: It is unclear what the authors exactly mean by 'the extension of aerosol'. Is it the vertical extension, or does it also include aerosol optical thickness?

Reply: We did refer to the vertical extension and therefore changed the text to "changing the vertical distribution of aerosol and NO₂ layers".

Comment: P3229, I26-29: In my view, it is not the purpose of a NO₂ retrieval algorithm to identify the type and distribution of aerosols. NO₂ retrievals are necessarily concerned with the ultimate effect of aerosols on TOA radiances and on light paths, in order to minimize the mismatch between the observed and (forward modelled) simulated TOA radiances/lightpaths (i.e. AMFs).

Reply: The sentence in question here was leading to confusion and misinterpretation of the message that we wanted to pass and therefore we have deleted it from the manuscript.

Comment: P3230, section 2.1: Could you please provide some more details on SCIA-C1306

TRAN? Was polarization included? What pressure, temperature grid (vertical spacing) has been used? What is the source of the p,T information?

Reply: The version 2.2. of SCIATRAN does not include polarization and therefore this was not considered in the present study. We have added this information and the further requests in the text.

Comment: P3231, section 2.2: I suggest the authors point out that CHIMERE's model top does not extend above 6.5 km. Could the authors say some more about whether CHIMERE NO₂ profiles have been evaluated against profile observations? After all, the authors consider CHIMERE profiles to be 'more realistic'.

Reply: This has been changed and it was also noticed that 6.5km was a typo (and therefore corrected) as the model top goes to 500hPa which is approx. 5.5km.

Comment: P3237, I22-23: My suggestion is here that the authors specify their statement such that 'if the effect of scattering by aerosols are not accounted for in the retrieval, the NO₂ VC will be overestimated'.

Reply: Agreed and changed.

Comment: P3238, I12: 'when effects of aerosols are neglected in the retrieval'.

Reply: Changed to "when effects caused by aerosol presence are neglected in the retrieval".

Comment: P3239, I23-25: that the aerosol effect appears small here, appears to be at least partly due to the low single scattering albedo (0.87) in scenario J. This should be mentioned as well.

Reply: In Fig 11 from the original manuscript the same scenario was used for the SSA case study. As it is possible to see, the different SSA values have a small impact on the AMF values (about 20% on average) for this specific situation. This led us to conclude that the SSA value is in fact not so important for the results here (because in other

cases the different SSA has resulted in much higher variations). We have also tested two other situations: NO₂ profile to 1.0km height with this aerosol profile from scenario J; and NO₂ urban profile from CHIMERE with a box aerosol layer of 1.0km. From this we found that the change in the NO₂ profile has almost no impact on the AMF but major differences are found for the urban NO₂ with box aerosol (presented in Fig 6 from the original manuscript). Therefore we concluded that the main factor in this scenario was the aerosol vertical distribution rather than the properties of the particles.

Comment: In section 3.3, it is argued that the albedo and shielding effect cancel for situations with aerosol mixed with NO₂. It seems that the AOD does not really affect this cancelling, but what about the single scattering albedo? Based on later results, would you expect shielding to become dominant over the albedo effect for lower single scattering albedos?

Reply: This is a good point and we thank to the referee for bringing that to our attention. It is true that for this case the AOD seems not to have an influence on the results. However, we have looked further into our calculations and found that in the case of highly absorbing aerosol (low SSA) variations of the AODs will result in bigger differences in the AMFs that were not found before: the AMFs decreases for higher AOD by about 14%. The case mentioned in section 3.3 (in the original manuscript) where a cancelling of enhancement and shielding is found refers to scenario D (aerosol layer from 0.6 to 1.0 km). For this scenario we did find that the AMFs become much smaller when the aerosol is more absorbing than initially set. So for this situation, the shielding effect of the trace gas will be more pronounced than a potential enhancement of the NO₂. In the text we have added: "It should be noted though, that this is not the case for lower SSA. In the presence of highly absorbing aerosol, the shielding effect will be dominant and a decrease of the AMF is found. Therefore, the cancelling verified for the case presented here is related to the definition of the aerosol properties." In addition, it is also interesting to see the case of scenario F (aerosol layer up to 2.0 km) where the initially observed enhancement effect from the aerosols is now, with SSA set to 0.80,

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the opposite, i.e., the AMFs are smaller than the case without aerosol.

Comment: P3242, I17, there seems to be a redundant comma after 'both'.

Reply: Corrected.

Comment: P3242, I19-20: this sentence appears a little cryptic. In fact you say everything you want to say in the two following sentences.

Reply: We agree that the same information was sort of repeated in the mentioned sentences. Therefore, the text was changed to "In addition, differences in the air mass factors were found when applying either coarse or fine aerosol size distribution. However, no large differences were evident when considering small variations of those main types."

Comment: P3243, I7: suggest to reformulate 'If the scattering effects of an elevated aerosol layer are not accounted for, . . .'

Reply: We do not see the point of the referee here – the sensitivity reduction is independent of whether or not one accounts for the aerosols as it is a direct result of radiative transfer. It can be compensated for but still less photons will probe the NO₂ layer.

Comment: P3243, I22: what is exactly meant here by 'largely extended'?

Reply: The "largely extended" expression was replaced by "aerosol layers that expand to higher altitudes in the atmosphere".

Comment: P3244, I8-18: Since a good number of retrievals corrects for the effects of clouds, I think it would be appropriate make the call here for a more thorough investigation of the impact of aerosols on cloud retrievals in clear-sky and partly cloudy conditions.

Reply: We agree and have taken up this point in the text.

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