

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 for his/her useful comments which helped to improve on the structure of the manuscript and also on the technical and scientific content. In the following text we will answer all the comments and questions point by point.

General comments:

Comment: 1) The paper analyses the impact of several aerosol parameters on AMFs, i.e., a multidimensional parameter space, which is of course challenging to break down to structured results and conclusion. Nevertheless, the overall structure of the paper

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might be improved. In detail, I suggest to introduce some additional sub-sections, especially in section 2.3 (one sub-sections per parameter, in analogy to section 3). Also the different approaches (A to H versus I to P) could be separated by new sub-sections.

Reply: Thank you for this advice which certainly helped to improve the manuscript. Following these advices we have restructured the paper. The suggested changes combined with the revisions requested by referee 1 were performed and the final manuscript is now structured in more sub-sections according to the different topics addressed.

Comment: 2) In this study, the sensitivity of satellite observations for tropospheric columns is expressed in terms of airmass factors (AMFs). Given that AMFs are the central quantity, their definition is far too vague. In particular, no clear distinction between the height dependent sensitivity (Averaging Kernel or box-AMF) and the total AMF is made; the former does not depend on the (relative) trace gas profile, but the latter does! Both quantities are inaccurately mixed in this study using the same term (Fig. 1 shows box-AMFs!). I thus recommend to add an extra paragraph (like "The concept of AMFs"), preferably at the beginning of the method section, to clarify this matter, and give also some equations.

Reply: The reviewer is right that the distinction between total AMF; block AMF and averaging kernel was not made clear enough in the paper and we have tried to improve on it by a) adding a short introduction to the airmass factor concept including some basic equations and b) making sure that the nomenclature is used in a consistent way.

Comment: 3) Only one (quite low) value of 0.03 was considered for the surface albedo. This is not sufficient: even if this value would be representative for urban areas, NO₂ plumes might also be transported out of the cities; in addition, there are sources at non-urban sites, e.g. power plants, or biomass burning. The authors have to repeat their study for at least one higher surface albedo value, and report on the resulting

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effects.

Reply: Thank you for this suggestion. We initially thought that including results for different surface albedo could result into a diversion of the scope of the paper. We have now added the results for runs with surface albedo of 0.01, 0.07 and 0.1. This setting is certainly important for the determination of the NO₂ AMF and the results show that good knowledge of surface albedo is an important prerequisite for accurate retrievals.

Comment: 4) Aerosols also have effects on cloud retrievals. This aspect is shortly mentioned in the conclusions, but should be discussed before in some more detail, at least qualitatively. To what extent do cloud correction schemes also correct for aerosol effects, and under which circumstances clouds and aerosols have to be treated differently?

Reply: We agree with the reviewer that clouds are very important for the retrieval of tropospheric NO₂ columns and that this point was not given enough attention in the original text. In response to this comment and the suggestions made by the other reviewer, we have added a discussion on cloud effects to the manuscript. However, a quantitative analysis of all the possible scenarios with clouds, aerosols and the different approaches taken to correct for their effects will have to be the subject of a future study.

Comment: 5) The impact of aerosols is the main topic of this study, but figs. 5-11 show AMFs as function of the SZA. One can not see the general effect of AOD on AMF easily. I strongly recommend to reorganize the figures in such a way that the aerosol property under investigation is presented on the x-axis (either replacing or supplementing the existing figures). On the other hand, some differentiations of the plots might be left out (e.g., the discrimination of DD and BB in fig. 8, which has no effect) and just be mentioned in the text.

Reply: We agree with the referee that presenting the figures with AMF as function of SZA does not directly show the variation of the AMF caused by the parameter in focus.

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Still, this was the best way we found to present such results because, first, this is an important and varying parameter in the satellite measurements and, second, because this is a way to keep the same structure of figures throughout the case studies analysed. In response to the reviewer's comment we have improved the figures by adding more graphs in Fig. 5 and 6 (from the original version) showing also the variation of AMF as a function of AOD or SSA for a selected SZA (50°).

Minor comments:

Comment: 1) The introduction is extensive and might be shortened. Subsections 1.1 and 1.2 are partly rather "methods" than "introduction".

Reply: Following this advice and previous ones we have changed the text with section 1.1 and 1.2 now being mostly part of the section 2. Methodology.

Comment: 2) p3222 line 3: rather "substantially" than "partly"

Reply: The word partly was removed.

Comment: 3) p3222 line 17: delete "a"

Reply: Corrected.

Comment: 4) p3223 line 8: "interesting information": please be more specific.

Reply: The expression "interesting information on aerosols" was replaced by "information on aerosols (mainly aerosol optical depth (AOD) but also, e.g., aerosol size distribution)".

Comment: 5) p3226 line 20: how far is this "a first approx."?

Reply: The intensity weighting approximation used in this argument is only valid in the case of optically thin absorbers. In most cases, NO₂ absorption is small enough to make this a very good approximation, and only in cases of heavy pollution, deviations are expected. We therefore have replaced "first approximation" by "For an optically thin

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absorber, the overall absorption signal is determined by”.

Comment: 6) p3231 line 3: "0.6, 1.0 or 2.0 km height".

Reply: Corrected.

Comment: 7) p3231 line 25: "datasets of"

Reply: Corrected.

Comment: 8) p3233 line 12: In heavily polluted scenes, AOD may be considerably higher than 0.9.

Reply: The referee raised a good point and we definitely agree that much higher AODs can be found in some regions or for specific events. In fact, in the conclusion we mention that in the sentence: "Nonetheless, situations of highly polluted scenes, as those of megacities, were not exactly represented here (AOD can be much higher than 0.9)." We have here defined it as heavily polluted mostly in comparison to the other values used in the study. Still, to follow the advice of the referee and avoid misinterpretation of the definitions we have deleted "heavily".

Comment: 9) p3233 lines 17-21: In line 17, SSA is varied between 0.8 and 0.95, but in line 20, it is varied between 0.93 and 1. More confusing, the latter range (where the minimum is almost equal to the maximum of the former range) is used to estimate the "maximum effect".

Reply: Thank you for this comment which made us notice that the explanation of what was exactly done was not clear. For all the runs performed for the scenarios with the aerosol box profiles the AMFs were calculated for aerosol with SSA=0.93 and purely scattering aerosol (SSA=1.0). The 0.93 value was selected in the very beginning of this analysis because this was the average from the Dubovik et al. (2002) paper from where most of the aerosol properties needed for this study were taken from. The SSA=1 was used to determine the maximum effect in the AMF of the scattering properties of the aerosols. Later on we decided to do also this case study where we could analyse

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specifically the impact of variations in the SSA. Here we selected the 0.8 and 0.95 as the minimum and maximum values found for one of the measured profiles selected where the SSA was given for different heights of the aerosol layer. The text was now changed and hopefully a better explanation is given so that the reader can understand what was done and why we have selected those values. In addition, in the Fig. 11 (of the original manuscript) the results for SSA=0.93 were left out to avoid confusion.

Comment: 10) p3235 lines 9-11: If the profiles are not representative, but extreme scenarios instead, they might indeed have impact on the conclusions!

Reply: We certainly agree. Representativeness was not the correct word to use in this context and we have replaced it by accuracy as this was actually what we meant in the first place.

Comment: 11) Results: The labels used for the scenarios (table 1) should be added in the description of the scenarios in the plain text (for instance p3237, lines 6-9).

Reply: The scenario labels A, B, C, etc were added in the text according to the suggestion of the referee.

Comment: 12) p3237 line 11: correct "a too low the BL".

Reply: Corrected.

Comment: 13) p3238 lines 10-12: Is this also true for low SSA?

Reply: When low SSA is considered the conclusions are not the same. For the majority of the case scenarios it was found that aerosol mixed with the trace gas would result in enhancement of the AMF. This is not true if highly absorbing aerosol is present, e.g. with SSA=0.80. In fact, the previous enhancement effect is now dominated by a shielding of the trace gas and, therefore, the AMFs of the no aerosol scenario are higher than those with aerosol. We had missed to point this out in the text and added now extra remarks to remind that the conclusions mentioned refer mostly to the SSA value considered and that for more absorbing aerosol this will change.

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Comment: 14) How far is an aerosol layer above the NO₂ layer different from a cloud? (see (4) of the general comments)

Reply: The referee is right that aerosol layers in general behave quite similar to a thin cloud, in particular if they have large SSA. In the case of an aerosol layer above the NO₂, the similarity is even more pronounced, and in fact remote sensing measurements are often unable to distinguish between the two situations. The main difference we see is the expected optical thickness and the absorbing properties which will mainly have an effect on the behaviour of cloud correction algorithms.

Comment: 15) Section 3.4: Again, the values for SSA are confusing. Why not have just 3 values (0.8, 0.9, 1) in the RTM runs to get the general picture?

Reply: The values 0.8, 0.9 and 1 would have provided also a general picture of the variations. We did not perform all the runs with three SSA values because that would be very time consuming. Therefore, as explained above, we started the runs with SSA=0.93 taken from Dubovik et al. (2002) and SSA=1.0 and only for specific scenarios we have performed the case study of SSA variation.

Comment: 16) p3243 lines 1-3: Even without aerosols, an underestimation of the BL leads to overestimation of NO₂ the column!

Reply: This is certainly true and the point was not made clear enough in the original manuscript. The following text was now added in this section: "Before discussing the impact of the boundary layer height with trace gas and aerosol mixed in the atmosphere it is important to mention that the variations of the boundary layer influence the AMF calculations even when considering only a layer of NO₂ without aerosol present. When the top of the NO₂ layer expands from 0.6 km to 2.0 km the AMF will increase on average by a factor of 1.4. This is related to the fact that the sensitivity of the measurements is smaller close to the surface."

Comment: 17) p3243 lines 4-5: Is this also true for low SSA?

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Reply: One of the general conclusions regards the enhancement of the measurements when aerosol is mixed with the trace gas. We had initially written that "Aerosol mixed with the trace gas, even if only partly, will, by means of increased effective albedo and multiple scattering, enhance the NO₂ signal." However, this holds only for those cases where SSA=0.93 and the opposite is found when we consider lower SSA values. Therefore, because this question was raised several times in the review we have added the following sentence in the conclusion section: "Still, it is important to mention that these findings hold for the SSA considered here (0.93) and that a dominant shielding effect is found in the event of highly absorbing aerosol mixed with the NO₂."

Comment: 18) p3243 line 26: "moderate": please be more specific.

Reply: In the text we have added: "...are moderate because the AMFs vary only by ~7% on average."

Comment: 19) Given the importance of simultaneous profile information of both aerosols and NO₂, it might be worth adding a reference to the potential of MAX-DOAS measurements.

Reply: This is a good point and we have added a statement on the possible use of MAX-DOAS to the text.

Comment: 20) The conclusions miss some important aspects out: What is the conclusion for operational NO₂ retrievals? Can you give an estimate of the errors made by neglecting aerosol effects (or considering them being corrected by the cloud retrieval) in current NO₂ retrievals (at least a sign)? Do you recommend to ignore aerosol effects until better aerosol data is available? Which kind of aerosol information (AOD, BL, ...) is needed (with which accuracy) to significantly improve the NO₂ retrieval, and is there a chance to have it in near future?

Reply: These points are certainly important and relevant for the study presented here. Unfortunately, we cannot answer to all the questions raised. Considering the high vari-

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ability found in the results it is difficult to estimate the errors made by neglecting the aerosol effect. This error is dependent on several factors and it was shown that in some situations the variations on the AMFs are almost zero while in other events it can be close to a factor of 2. In the conclusion we have identified aerosol parameters (AOD, SSA and distribution) and NO₂ vertical distribution as important factors in the calculations that need to be included and/or improved in the retrieval of tropospheric columns. In addition we have also written a short discussion on potential future solutions and recommendations for future work for the improvement of what is currently done. We do believe that with the data available currently it is possible to work towards a better NO₂ product. Satellite aerosol products have their own caveats and at the moment it is hard to find a dataset that would provide all necessary information with high accuracy and required resolution. Still, this data is helpful, provides valuable information and integration of datasets can be attempted.

Interactive comment on Atmos. Meas. Tech. Discuss., 2, 3221, 2009.