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Atmospheric Measurement Techniques Discussions

Interactive comment on "Space-based retrieval of NO_2 over biomass burning regions: quantifying and reducing uncertainties" *by* N. Bousserez

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Response to anonymous Referee #2:

I would like to thank the anonymous referee for his useful comments and suggestions about the manucript. Please find my responses to each of them below, as well as a revised manuscript attached (pdf file).

General comments: Some parts of the manuscript by N. Bousserez would provide a worthwhile contribution to the scientific literature on the relevant topic of aerosol corrections for trace gas retrievals. For instance, the contrasting impact of aerosols on the clear-sky air mass factor over Canada (slight increase in NO2 air mass factor) versus over Africa (decrease in NO2 air mass factor), is interesting, and can in principle be well

understood given the similar vertical distributions of aerosols and NO2 over Canada ('albedo effect'), compared to the elevated aerosol layer above the biomass burning NO2 over Africa ('screening effect'). These parts are useful, and, to my knowledge, have not been studied before. However, after these relevant sections, the manuscript derails altogether because it claims to provide an aerosol correction for current, operational NO2 retrievals, without actually staying consistent with those retrievals. The major weakness is that the author makes the implicit assumption that the effect of aerosols on the cloud parameters is negligible. However, the source of information on cloud characteristics, are the satellite measurements themselves, and these are known to be sensitive to the presence of aerosols. Furthermore, there are far too many technical and scientific errors in the manuscript, and I found fact-checking and appropriate referencing to be unusually sloppy. Below I specify all my concerns (they mostly come down to one and the same thing) and suggestions on how to address them. In my opinion this manuscript should be rejected, but may be suitable for publication once the author solves the most pressing scientific issue.

Response: In the revised manusript the problem related to the perturbation of the retrieved cloud parameters by aerosols has been clarified and the results reanalyzed (see responses below). Note that in this study we do not propose a correction for the aerosol effects, but only for the shape factor errors. Only the effect of aerosols on the AMF , and its sensitivity to several retrieval parameters is investigated here. Also, the fact that the cloud correction cannot implicitly correct for the aerosols effect when real clouds are present has already been explained in Leitao et al. (2010). Results from previous studies by Boersma et al. (2004, 2011) demonstrated the existence of a (partial) implicit aerosol correction for clear-sky cases only. In our study we reexamine the case of cloudy scenes. Although we do not simulate the effect of aerosols on the retrieved cloud parameters, we clarified why the aerosol sensitivity study provides valuable results to improve our understanding of the cloud/aerosol retriesval problem.

Major concerns 1) The most important claim in this manuscript, i.e. that the effect of

aerosols on the NO2 air mass factor (AMF) is not fully taken into account by modified satellite-observed cloud parameters, remains unproven. The trouble starts with the definition of the 'total biomass burning AMF correction factor' on page 6652-6653. This definition is only correct if the author takes into account the effect of aerosols on the retrieved cloud parameters. On page 6649, the author recognizes that the 'AMF formulation we use takes into account cloud-contaminated pixels, as described in Martin et al. (2002)'. In the presence of aerosols, the OMI O2-O2 algorithm generally returns higher cloud fractions and higher cloud pressures, although the magnitude of the impact depends on the type of aerosol. The numerator in the aero_cor (Eq. 3), AMFaero_bb, should take this dependency into account, and it is obvious that the author has not done so. He should stay consistent with the retrieval framework he builds on, by calculating both the old and new AMFs taking into account (modified) cloud parameters as described by Martin et al. [2012].

Response: The total biomass burning correction in Section 5 (which included both aerosol and shape factor effects) has been replaced by a shape factor correction that represents the effect of the fires-modified NO2 shape factors on the AMF, and does not include aerosol effects. Therefore in this case the effect of biomass burning aerosols is assumed to be implicitly accounted for by clouds. A restrictive cloud filtering (cloud fraction < 5%) is used in order to limit the impact of real clouds on the retrieval.

2)The author fails to live up to his promise "We analyse the interplay between clouds and aerosols in the algorithm" (P6648). He studies the one-way effect of clouds on the aerosol correction, but the impact of aerosols on the cloud parameters is not accounted for. Figure 9 is an illustration of this flaw. The figure claims to show the aerosol correction as a function of 'cloud irradiance fraction'. In the Figure, the author assumes a so-called 'pre-existing' cloud, which bears no relation whatsoever to a cloud fraction and cloud pressure retrieved from measurements by OMI or any other UV/Vis nadir instrument. Because the widely used O2-O2 (and FRESCO) algorithm return modified cloud fractions and cloud pressures in the presence of aerosols, the concept of the 'pre

existing cloud' provides a distorted view of the true effect of aerosols on the trace gas retrieval. The author should therefore feed his aerosol scenarios to a realistic cloud retrieval (model) and evaluate the changes in the retrieved cloud fraction and cloud pressure. Subsequently, he should be consistent and calculate the effects of these modified cloud parameters in his AMFs, which 'takes into account cloud-contaminated pixels, as described in Martin et al. (2002)'. Without such a step, this manuscript remains nothing but a brief exercise in sensitivities for only a part of the retrieval concept (i.e. the 'clearsky' AMFs), and therefore not representative for the retrieval framework as a whole. Lin et al., ACPD, 2013 take first steps to such an approach. Lin, J.-T., R. V. Martin, K. F. Boersma, M. Sneep, P. Stammes, R. Spurr, P. Wang, M. Van Roozendael, K. Clémer, and H. Irie, Retrieving tropospheric nitrogen dioxide over China from the Ozone Monitoring Instrument: effects of aerosols, surface reflectance anisotropy and vertical profile of nitrogen dioxide, Atmos. Chem. Phys. Discuss., 13, 21203-21257, doi:10.5194/acpd-13-21203-2013, 2013.

Response: It is correct that the sensitivity study performed characterizes the effect of clouds on the aerosol correction, but does not quantify the impact of aerosols on the cloud parameters retrieval. This latter calculation, which would require to use the FRESCO algorithm (for instance) to reproduce the cloud retrieval, is beyond the scope of the present paper. The aerosol correction sensitivity experiment presented Figure 9 is intended to provide a theoretical understanding of how the aerosol effect is modified by the presence of clouds in the retrieval. From a realistic retrieval perspective, it is true that the presence of aerosols would, in principle, induce a minimum value for the retrieved cloud fraction, although here we consider all possible values from 0 to 0.3. However this does not impact the sensitiviy analysis, whose aim is to quantify the derivative of the aerosol correction with respect to cloud fraction.

3) Because the author did not stay consistent with his AMF formulation, misleading statements arise. One example is on page 6656, which states that 'the aerosol correction increases linearly with cloud irradiance fraction'. Apart from the erroneous term

'cloud irradiance fraction', the effect is exactly opposite to what can be expected based on the physics of the retrieval: the higher the cloud fraction, the more likely they are to outshine any aerosol effects. Bright white clouds enhance the TOA radiance levels in a much stronger way than aerosols.

Response: There is a misunderstanding of our statement here. What is said is that for a given aerosol scenario, increasing the cloud fraction results in an increase of the aerosol correction factor. Although the shielding effect of clouds does reduce the absolute effect of aerosols, the relative change in the AMF due to the presence of aerosol is greater when clouds are present.

4) Section 7 is particularly confusing and jumps to conclusions that are at best unproven, and most likely false. The author mentions the 'particular case' of "pre-existing clouds in a scene". According to the author, such pre-existing clouds would modify the effect of aerosols on the AMF. However, the author assumes here (without telling us so) that a pre-existing cloud (i.e. a satellite-observed fraction, pressure) is insensitive to the presence of aerosols. I strongly dispute the implicit assumption that satellite retrieved cloud parameters are insensitive to the presence of aerosols. If the author thinks they are, he should prove it by means of a sensitivity study with a realistic cloud retrieval in response to cases with (a) a pre-existing cloud of certain pressure and fraction, and (b) the same pre-existing cloud in combination with the aerosols as assumed by the author to berepresentative of biomass burning aerosols.

Response: The term "pre-existing clouds" has been replace by "clouds", since it is a source of confusion. The fact that in the presence of clouds the aerosol effect cannot be implicitly accounted for has already been suggested by Leitao et al. (2010). It stems from the fact that the presence of surface scattering aerosols will increase the retrieved cloud fraction while not modifying the cloud top pressure (when clouds are above the aerosol layer). This will result in an overestimate of the cloud shielding effect while the aerosol-driven sensitivity increase at the surface is not simulated. Regarding the need for a cloud retrieval sensitivity study, please refer to response 2.

5) On page 6657, the statement that "this aerosol correction perturbation is not associated with any perturbation of the retrieved cloud parameters (it is an artifact of clouds)" plainly contradicts evidence given in the peer-reviewed literature that cloud parameters are perturbed by aerosols (Boersma-papers from 2004 and 2011). The statement is not backed up by any observations or simulations. The statement, and the underlying idea that clouds somehow appear ex machina (or be pre-existing), is at odds with the retrieval concept using the independent pixel approximation followed by the Dalhousie, andKNMI, and NASA retrievals, and also by the author.

Response: The statement "this aerosol correction perturbation is not associated with any perturbation of the retrieved cloud parameters (it is an artifact of clouds)" has been removed. We were referring to the effect of clouds on the aerosol correction. We now explain why explicitly taking into account the effect of aerosol may be beneficial in the presence of clouds. Note that the fact that the cloud parameters are modified by the presence of aerosols is not contested here, but only the fact that these modifications can implicitly account for the impact of aerosols when clouds are present, which is also suggested by Leitao et al. (2010). Please see Section 7 of the revised manuscript for more details.

6) Section 9 presents an interesting idea, but a discussion about the use and applicability of this approach for other biomass burning regions and times is missing altogether.

Response: After reexamination, we believe the proposed formula has actually a greater generality than first stated. The derived formula could in principle be used as a near-real time correction to the retrieved NO2 tropospheric column over any NOx source. Since the aircraft campaign data used for this study do not allow to validate the proposed correction, future work will consist in evaluating this method using ground-based or dedicated aircraft campaigns. This is now explained in detail in Section 9 of the revised manuscript.

Other concerns and technical suggestions:

(T1) The distinction between tropospheric and stratospheric contributions to the total NO2 column is not being made on page 6649. Furthermore it is unclear what integration of eta from 0 to 1 means in Eq. 1.

Response: The distinction between tropospheric and stratospheric contribution in the total NO2 column is now made in this Section. Also, the definition of eta has been added.

(T2) Awkward: 'Vertical sensitivity of 'radiance' observed by the instrument to NO2'. The radiance itself can hardly be described as sensitive to NO2. A NO2 signature can be identified in the reflectance spectrum. Such signatures are generally stronger if the NO2 resides at higher altitude.

Response: 'Vertical sensitivity of 'radiance' observed by the instrument to NO2' has been replaced by 'the sensitivity of the backscattered spectrum to the abundance of NO2 at each sigma-level.'

(S1) Suggest to merges Figures 1 and 2. They provide too limited information to justify them a stand-alone figures.

Response: Figure 1 and 2 have been merged.

(S2) Suggest to merge Figures 3 and 4 and group them in a similar way as done in Fig. 5. These figures should have a legend to quickly see what is represented by the symbols. Such a legend is missing now.

Response: Figure 3 and 4 have been merged and a legend now describes what is represented by the solid and dashed lines.

(S3) It is completely unclear what a 'scattering profile shape' is. Apparently 'scattering' is unitless, and cannot only be observed, but also modelled. Obviously this should be explained.

Response: The unit (km-1) has been added. In Section 4, we now provide a reference

(Martin et al., 2003) explaining how the aerosol optical properties are simulated in the model.

(S4) On page 6653, and in the caption of Fig. 5 the 'cloud irradiance fraction' is mentioned but this is very sloppy. A cloud irradiance fraction does not exist, and the author probably intended to refer to the cloud radiance fraction. Various papers in the peer reviewed literature use the concept of the cloud radiance fraction, and these should be cited.

Response: "Cloud irradiance fraction" has been replaced by "cloud fraction" throughout the manuscript, a term used in Martin et al. (2003).

(S5) Figure 6 is awkward since the y-axis is much more compressed in the 'Africa' plot than in the 'Canada' plot.

Response: This figure has been removed, following another reviewer's suggestion.

Specific comments:

P6647, L10-11: The author calls the NO2 retrieval a "two-step process", but the stratospheric correction is an important third step that is not even being mentioned.

Response: The step that derives the tropospheric part of the slant column is now described.

P6647, L17: Russell instead of Russel. Response: This has been corrected throughout the manuscript.

P6647, L19-21: many CTMs resolve temporal and spatial patterns of biomass burning emissions to some extent. For instance the widely used GFED-2 provides 8-day averaged emission factors. To say that high spatial and temporal variability is generally unresolved is too strong, and should be rephrased as 'not fully resolved'. This comment also applies to P6652, L23-24, which should be nuanced as well. Response: This statement has been nuanced according to the reviewer's suggestion.

P6649, L5-6: please be explicit and describe this as the independent pixel approximation. Response: This has been added to the text.

P6649, L18: typo 'aicraft'. Response: Corrected.

P6651: although it is mentioned in the appendix, it is important enough to describe within the main text of the manuscript which spatial and temporal resolution the GEOS-Chem simulations had. Response: The spatial resolution of the grid as well as the temporal resolution of the biomass burning inventory are now specified.

P6652, L19-21: this conclusion is too strong. The comparison at best suggests that GEOS-Chem provides a reasonable first order simulation of aerosol and NO2 properties. There are still many things unclear however, such as the spatial extent of the 2 x 2.5 grid cell viz-a-viz the spatial representativeness of the aircraft measurement, and the temporal representativeness as well. This part should be nuanced. Response: We replaced the last sentence by:" In conclusion, the evaluation of the GEOS-Chem NO2 profiles and aerosol optical properties over boreal and savanna fires suggests the model provides a reasonable representation of their main characteristics. In the following, aerosol and NO2 profiles simulated by GEOS-Chem are used to calculate the AMF and analyze its sensitivity to biomass burning emissions."

P6653, L13-15: mention what the source of information was for the cloud radiance fraction, and cite accordingly.

P6653, L16-21: the statement that 'aerosols increase the AMF' holds because the vertical distribution of the aerosols is similar to that of NO2 for the boreal fires. That specification should be made for clarity. Response: In order to avoid the problem related to the implicit aerosol correction by clouds and to simplify the manuscript (following another reviewer's advice), now only the shape factor-related AMF correction is shown and discussed in this Section.

P6654, L7: 'narrower'! shallower. Response: Figure 6 has been removed following

another reviewer's suggestion and the corresponding sentence too.

P6654, L22-23: it is completely unclear how the selection that minimizes 'the representativeness error of NO2 profiles' was made. What is meant with chosing those profiles with the highest fire emissions? Response: Each simulated NO2 profile corresponds to the average of the NO2 profiles over a 2x2.5 grid cell, themselves representative of the average of the underlying NOx emissions over that area. The OMI pixels considered here having a horizontal resolution of approximately 0.1x0.1, the NO2 profile representative of the retrieved scenes over fires will correspond to higher NOx emissions compared to the GEOS-Chem grid. Therefore, considering the higher end of the emission spectra for GEOS-Chem helps matching more closely the NO2 profiles "observed" at the OMI pixel resolution.

P6654, L25-26: 'with and without the elevated aerosol layer': it is unclear what the source of information is for this aerosol layer . . . was it simulated by GEOS-Chem? Has it been inferred from the CALIPSO observations? Response: In the second sentence of Section 6, it is now specified that both the NO2 and aerosol profiles are obtained from a GEOS-Chem simulation: "A sensitivity analysis is conducted using two reference aerosol and NO2 profiles from the GEOS-Chem simulation during DABEX."

P6655, L3-9 and 15-17: The sensitivity of aerosol correction to the single scattering albedo shown in Figure 8 is pretty similar as found by Boersma et al. [2004]. It would be appropriate to refer to and discuss the SSA-dependency results in that perspective. Response: The aerosol correction sensitivity to SSA shown in Fig. 9 of Boersma et al. (2004) shows a increase of the correction factor from w0=0.88 to w0=0.96 (consistent with our finding), but a decrease of the correction factor from w0=0.88 to w0=0.89. This behavior is in contradiction with our results, and it is unclear how it is possible to decrease the aerosol correction factor by increasing the SSA.

P6656, L16-20: The message in the Boersma-papers is that correcting AMFs for aerosols cannot be decoupled from correcting cloud retrieval schemes for aerosols.

The text should be nuanced accordingly. Response: This part has been nuanced.

P6657, L13: for DOMINO, a cloud fraction of 0.3 is certainly not the threshold used. The flagging occurs for cloud radiance fractions > 0.5, and this is consistent with cloud fractions higher than 0.15-0.20. Response: The statement about the cloud fraction threshold used in operational DOMINO retrievals has been removed.

P6658, L5-6: the author should be precise here. Which operational retrieval uses climatological NO2 profiles, and what is exactly climatological about those profiles? Response: The OMI NO2 product (NASA standard product) is now explicitly mentioned and the origin of the NO2 climatological profile used explained.

P6658, L17-18: the Eskes et al. reference is missing from the reference list. Response: The reference has been added to the list.

P6658, L21-23: there appears to be no difference between simulations A and B. Response: A: corresponds to a pseudo-retrieval of NO2 columns from a GEOS-Chem simulation that includes biomass burning emissions, but using a NO2 shape factor from a simulation without biomass burning in the AMF. B: corresponds to a pseudo-retrieval of NO2 columns from a GEOS-Chem simulation that does not include biomass burning emissions, and uses a NO2 shape factor from the same simulation (without biomass burning) in the AMF.

P6659, L5: the relationship between delta NO2 here and in L1 should be made clear. Response: The derivation of this equality is now detailed.

P6659, L6: it is completely unclear how Figure 11 has been generated. Does the x-axis represent the delta NO2 from line 5 (observed) or line 1 (simulated)? Response: See previous response. The delta NO2 defined in L1 and L5 are the same.

P6660, L9: I think the author should refer to Fig. 11 instead of Fig. 12 here. Response: This has been corrected.

P6661, L15-18: it is unclear what the author has in mind here. Response: The sen-

tence the reviewer refers to has been clarified: "It was found that the intensity of fire emissions does not significantly impact the effect of aerosol for a given NO2/aerosols emission ratio. This suggests that emissions-related representativeness errors may be weak when modeling the aerosol effects."

P6661, L24-28: as said earlier, the authors should provide evidence for the statement that an “implicit cloud correction cannot fully account for an explicit aerosol correction”. Because the author has done nothing to evaluate to what extent cloud retrievals do pick up an aerosol signature, he cannot imply that the presence of pre-existing clouds requires an explicit aerosol correction. Response: As explained earlier in these responses, Leitao et al. (2010) already showed that in the presence of clouds above surface scattering aerosols, the modified cloud parameters cannot theoretically account for the aerosol effects, but in fact produce an opposite effect (shielding). Although no cloud algorithm sensitivy experiment has been performed in their study, their discussion is based on basic understanding of the physics of UV-VIS cloud retrievals. In our study we combine that idea with results from our sensitivity analysis to show that in this case applying an explicit aerosol correction should be beneficial to the retrieval.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/6/C4279/2014/amtd-6-C4279-2014-supplement.pdf

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