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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

Anonymous Referee #2

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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

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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.

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].

(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

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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 ‘clear-sky’ 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.

(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.

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(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 be representative of biomass burning aerosols.

(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, and KNMI, and NASA retrievals, and also by the author.

(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.

Other concerns and technical errors:

(T1) The distinction between tropospheric and stratospheric contributions to the total NO_2 column is not being made on page 6649. Furthermore it is unclear what integration of eta from 0 to 1 means in Eq. 1.

(T2) Awkward: 'Vertical sensitivity of 'radiance' observed by the instrument to NO_2'. The radiance itself can hardly be described as sensitive to NO_2. A NO_2 signature can

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be identified in the reflectance spectrum. Such signatures are generally stronger if the NO_2 resides at higher altitude.

Technical suggestions:

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

(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.

(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.

(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.

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

Specific comments:

P6647, L10-11: The author calls the NO_2 retrieval a "two-step process", but the stratospheric correction is an important third step that is not even being mentioned.

P6647, L17: Russell instead of Russel.

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

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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.

P6649, L5-6: please be explicit and describe this as the independent pixel approximation.

P6649, L18: typo ‘aicraft’.

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.

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.

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.

P6654, L7: ‘narrower’$\rightarrow$ shallower.

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?

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?

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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.

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.

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.

P6658, L5-6: the author should be precise here. Which operational retrieval uses climatological NO_2 profiles, and what is exactly climatological about those profiles?

P6658, L17-18: the Eskes et al. reference is missing from the reference list.

P6658, L21-23: there appears to be no difference between simulations A and B.

P6659, L5: the relationship between delta NO_2 here and in L1 should be made clear.

P6659, L6: it is completely unclear how Figure 11 has been generated. Does the x-axis represent the delta NO_2 from line 5 (observed) or line 1 (simulated)?

P6660, L9: I think the author should refer to Fig. 11 instead of Fig. 12 here.

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

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.

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 6645, 2013.