Atmos. Meas. Tech. Discuss., 6, C4291–C4302, 2014
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Atmospheric Measurement Techniques Discussions
Open Access

AMTD
6, C4291–C4302, 2014

Interactive Comment

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

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Received and published: 6 February 2014

Response to anonymous Referee #3:

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:

This paper deals with an important topic for the scientific community dealing with trace gases retrievals from satellite observations. It is a relevant subject and the paper can be a valuable contribution to the understanding of aerosol impact in the satellite retrievals.

To my knowledge, a study focused on satellite retrieval of NO2 from biomass burning events making use of measurements from specialized campaigns has not been presented before. The idea is interesting and has been discussed in previous publications so it is certainly a relevant subject and a novel achievement (if well conducted). There are many issues with the current form of the manuscript and as it is the paper cannot be published. Major revisions are necessary and my main concerns are raised below. The general structure of the paper is reasonable but the text becomes confusing at parts and often the author states facts not fully explained to the reader. A more complete exploration of scientific concepts with further depth sending across a clear message with ground-breaking conclusions is certainly missing from the manuscript. As a full revision is needed the conclusions should also later be made clearer and focused on what was actually found with this study and how can that be used in the future. The text needs some major revision on grammar and spelling. Some suggestions are made below but there is room for additional improvement regarding grammar and clear writing to help the reader follow the paper. For example, section 3.1 repeats too much information in different sentences and this could be more concise. The whole referenced literature needs a profound revision, not only regarding the reference list that does not include all those mentioned in the manuscript, as well as, addition of some more references on the topics explored such as: characteristics and emissions of biomass burning events (see below further comments); previous sensitivity studies done (several mentioned in Leitao et al. 2010 that is the only mentioned for the conclusions in P6655). Please provide references to facts stated such as: the campaigns mentioned in P6648; P6657 L19 the typical values of surface reflectances. There are many declared facts in the paper that are not supported with suitable literature. Missing references in the reference list are at least (but this was obviously not a comprehensive control): Logan 1983, Finlayson-Pitts and Pitts 1986, Leue et al 2001, Eskes et al 2003, Andreae and Merlet 2001. Verify as well the references in the text as sometimes errors are made in year Jacob et al. 2009 should in fact be 2010. Response: The manuscript has been corrected following the reviewer's suggestiong. In particular, grammar and spelling have been revised and the introduction has been updated with additional references (see below). Repetitions in section 3.1 have been removed.

Specific comments: 1) In the second paragraph of the introduction the author says the NO2 satellite retrieval is a two-step process. This would be the case if one considers the fit and the slant to vertical column conversion the two broad steps. However, in the latter there are different steps to be performed and it should be at least mentioned that the slant column determined from the fit is total and to obtain a tropospheric part several corrections are needed. Considering that the paper is mainly focused on the retrieval of NO2 and its uncertainties a proper description of retrieval process is essential.

Response: The retrieval step that derives the tropospheric slant column has been added to the retrieval description in the introduction.

2) Also, the paper fails to provide a comprehensive introduction of the problematic of satellite retrievals in the case of fires. What has been done in previous studies and how is the aerosol correction addressed by the different groups working on this? How is this correction useful and important for the scientific community? Furthermore it is important to make clear distinction from what is already done and what can be changed with suggested methodology of this study. Response: A more detailed description of the problematic of NO2 retrievals over fires has been added to the introduction. In particular, the potential magnitude of the aerosol effect from previous studies is now specified. Also, the fact that some groups consider the aerosol impact as implicitly accounted for by the cloud correction is emphasized. In the present study, we do not propose a correction for aerosols, but rather investigate their effect on the AMF and how clouds modulate it. It is now specified in what the proposed retrieval improvement differs from methods in previous studies. While most previous works relied on increasing the spatial resolution of the retrieval input parameters (e.g. the model simulation), in our study a measurement-based correction is introduced, which is intended to reduce the representativeness error in the simulated NO2 profiles.

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3) It is not mentioned anywhere in the manuscript that fire events are different in many characteristics (e.g., intensity and fuel and resulting type of emissions). Temporal and spatial variability are high but the different fire types also represent a challenge to accurately reproduce these events in models or apply standard correction to satellite retrievals. Again at this occasion a proper literature review support is missing. This should also be mentioned in the beginning of section 5. Response: The fact that NOx emissions from fires depend on several factors such as the type of vegetation, the burning phase and the combustion efficiency has been added in the 1rst paragraph of the introduction, as well as the corresponding references. This is also now mentioned in the beginning of section 5.

4) Also, in section 4 the measurements made are not put into perspective with previous findings and published studies that would support similarity or differences within events. For the AMF retrieval the retrieval of correct and typical optical properties and aerosol load is important and if the data from the campaigns is used as base of the study it is important to understand if the values are or not expected according to previous observations. Response: Comparison of the DABEX results with the previous SAFARI-2000 experiment is now mentioned in Section 3. Reference to a study by Reid et al. (2005) that supports the SSA values found for ARCTAS and GEOS-Chem is now included in Section 4.

5) In P6652 it is concluded that the higher values of the aerosol extinction coefficients are because of more intense fire emissions. It is not clear to me how the author came to this conclusion. Higher extinction does not necessarily and straightforward means higher emissions, as this factor depends on size, mass concentration and chemical composition of aerosol. From Fig. 6 with the different scales used for Canada and African events this conclusion cannot be easily perceived. In this study two very different fire types are analyse: boreal and savannah. It is true that the magnitude aerosol emission of each event might be quite different simply because of different burning phase and combustion efficient and fuel burned. But all these factors also influence

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the type of aerosol (mixture) found in each of the events, an issue not mentioned anywhere. This might be an important information for the study performed as, depending on the aerosol load, the different composition might lead to different impact on the retrieval and this issue is not address in the manuscript. Response: The comment on the different magnitude of the aerosol extinction over boreal and savanna fires has been removed. Indeed, as stated in this section, the poor sampling of the boundary layer during DABEX does not provide the statistical robustness needed to draw this conclusion. However, the results demonstrate that the model is capable of reproducing the main aerosol optical properties (which result from the different types of aerosol and their concentration), which are used in the AMF calculation.

6) Still in section 4, the AERONET data is compared to the DABEX campaign results. Why isn't this done for the other campaign as well? Why also in Fig. 6 the CALIPSO observations are only presented for DABEX? In fact, both campaigns are mentioned but then only one is fully explored. Response: There was no AERONET data available in the vicinity of the aicraft measurements for ARCTAS. Figure 6 has been removed, as suggested by another reviewer. DABEX has been chosen for the sensitivity experiment because the aerosol profile over African fires shows this interesting elevated aerosol layer structure, which allowed to test the sensitivity of the aerosol correction to different profile shapes.

7) In section 6 it is not clear what model is used to the sensitivity tests. The LIDORT model is mentioned in the abstract and section 2 but not in this section. Also it's not clearly explained what were the input parameters for the model and what info is taken from model and from measurements.

Response: This part has been clarified. It is now explicitly written that the LIDORT model is used for the AMF calculation and that the prior NO2 and aerosol profiles were generated using the GEOS-Chem model.

8)In P6656, L16 the sentence "Aerosol may impact the retrieval of cloud parameters."

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On its own and short like that it is a confusing sentence as the opposite happens as well, it all depends on what is to be retrieved from which instrument measurements. So please explain better what the idea is behind this sentence. Essentially, the relation and influence of aerosol on clouds and vice-versa is not clearly explained and becomes confusing to understand how the author managed to separate one from another. For example, in section 7 the concept of “pre-existing clouds” becomes very disconcerting. It is not clear what the author is referring to here and relation implied to what is used in the OMI retrieval. Furthermore it is not clear in the manuscript how the clear distinction is made (and found) between the effect of aerosols on the cloud retrieval and vice-versa (it is certainly a two-way relation but it’s not made understandable). To imply that one affects the other in the sensitivity study performed it is important to, in the first place, how aerosols affected the cloud parameters as well and, if this effect has been excluded state clearly how it was so. This is introduced at the end of section 6 but should be better explained how in fact it was possible.

Response: Section 7 has been entirely rewritten. The term "pre-existing clouds", which is confusing, has been replaced by "clouds". Also, we clarified the disctinction between the effect of aerosols on the cloud retrieval, and the effect of clouds on the aerosol correction. It is now explicitly stated that in this study we do not investigate the sensitivity of the retrieved cloud parameters to aerosols, but only the impact of the presence of real clouds on the aerosol correction. Although in practice aerosols will also perturb the retrieved cloud parameters, the merit of such an analysis is to provide a theoretical understanding of how the presence of clouds modifies the impact of aerosol on the AMF. We clarified and developed our interpretation of the results. In particular, in Section 7, we explain why the results suggest that in the presence of clouds taking into account the surface aerosols effect in the AMF should be beneficial to the retrieval.

9) In P6658, L10-11, it is said that NO_2 remains in the boundary layer only. Please provide a reference for such general statement or rephrase and focus on this particular case. What boundary layer height is assumed here? The full 2km where NO_2 is

present? In some events NO2 might in fact be transported to higher altitudes due to convection within the fire plume. It is not uncommon to find injection heights of smokes to the free troposphere. To my knowledge it is still not clear how high NO2 is injected and how this change across different events. Compared to aerosols and other trace gases the short life of NO2 complicates the analysis of this species in fire events. Response: We now provide the following references after our statement that "over high NOx sources [...] most of the NO2 column will be concentrated in the boundary layer": (Lamsal et al., 2008; Bucsela et al., 2008). Here we perform a statistical analysis to infer a relationship between slant column perturbations and shape factor corrections, based on simulated retrievals during DABEX. The boundary layer height is not fixed but varies between model grid cells. We do not simulate the possible effect of pyroconvection in our simulation. Although it is true that in some cases fire emissions can be injected into the upper troposphere, based on the profiles measured during the ARCTAS and DABEX campaign it seems this phenomenon did not play a significant role. 10)P6660, L11 the author says that one can see that uncertainties are smaller than those associated with missing biomass burning sources. At this stage is not clear what is being compared to what. At the end of the same section the author fails to mention and explore the potential of ground based measurements when it refers to only data from other aircraft campaigns. Campaigns provide very useful datasets but are still often limited to specific events with very particular characteristics.

Response: We clarified this paragraph, which is now part of Section 9, since Section 9.2 has been removed. The NO2 correction factor represented in Figure 8 of the revised manuscript can be interpreted as the retrieval error (factor) associated with missing biomass burning sources. The standard deviations shown can also be interpreted as the error made when using the proposed relationhip to infer the correction. It is now mentioned that in addition to aircraft campaign data, ground-based measurements should be used to evaluate the proposed retrieval correction.

11)The author claims to have found a correction method but it does not clearly eluci-

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date the readers if this method can actually be used on a near-real-time retrieval as automatic correction (and how would that be done) or just post-retrieval correction for specific cases. How would those cases be identified or selected? “In practice, this methodology could be extended to any type of NOx source, with possible adaptations of the formula over areas with very different retrieval characteristics (e.g. surface reflectance).” Can this study based on one particular event provide a solution with one formula that will solve the retrieval problems? These events are very different from one another, so is the solution found on this study applicable to other and all biomass burning events? What information or data is missing to adapt the correction to other events such as desert dust storms? Or peak urban pollution events? The idea presented is very interesting and important subject for progress in satellite retrieval but the author fails to properly explore and explain this.

Response: The statement "In practice, this methodology could be extended to any type of NOx source, with possible adaptations of the formula over areas with very different retrieval characteristics (e.g. surface reflectance)" has been modified. After reexamination of the result, we believe the proposed formula has actually a greater generality than first stated. Indeed the variability of the retrieval inputs (surface reflectance, aerosol shape profile) in the pseudo-retrieval experiment provides a reasonable sampling of different conditions, giving some confidence in the robustness of the derived relationship. The derived formula could in principle be used as a near-real time correction to the retrieved NO_2 tropospheric column over any source, as explained in Section 9. However, an experimental validation is necessary. Since the aircraft campaign data used for this study do not allow to validate the proposed correction, future work will consist in evaluating this new retrieval using ground-based or dedicated aircraft campaigns.

Technical corrections

1) It is awkward to read the whole paper as it is a group work (words “we” and “our” are often used) although there is just one author and no team collaboration is even ac-

knowledge. In general the paper would be better written in impersonal form. Response: The impersonal form has been used in the revised manuscript.

2) Suggestions for figures and tables: Combine table 1 and 2 as they provide the same information but for different campaigns. The same can also be done with Fig. 1 and 2 joined into one alone. (as in the text, the references mentioned in the tables are missing in the reference list) Response: The references in the table have been added to the reference list. Since slightly different measurement information is provided for each aircraft campain, the table have been kept separate. Figures 1 and 2 have been combined.

Figures 3 and 4 can also be combined, and a legend would help to identify which line is what. Maybe presenting the median and mean is not so useful (even if we all know that often these differ this difference is not really highlighted in the text). It is also not clear why for one case SSA is shown and for the other the scattering coefficients. Response: Figure 3 and 4 have been combined, and a legend now helps identifying which line is what. In the text (Section 4), it is now explained why scattering coefficient are shown for the DABEX campaign instead of SSA:" Since absorption measurements were available only at 565 nm, we scaled the observed extinction at 565 nm to 440 nm by applying the ratio between the scattering at 440 nm and 565 nm. In order to conserve original measurements in the comparison, here the choice has been made to show the scattering at 440 nm instead of the SSA."

Figure 5: the different colorscale in the aerosol correction does not help the comparison so please change that. You might also want to delete the extra "aerosol correction" in the figure for Canada. Response: The figures representing the aerosol correction and the total correction have been removed, since another reviewer pointed out the fact that the implicit aerosol correction through the modified retrieved cloud parameters was not considered. Only the AMF correction related to NO_2 shape factor perturbations is shown in the revised manuscript.

Figure 6: please change the figures so that the y axis has the same height for both Canada and Africa. Also use the same colorscale for the extinction coefficients. From this figure and with such lower values of extinction it's hard to understand how for the African fires the emissions are much higher. Maybe the AOD can also be calculated so that it can support the statement made in section 4. In the previous figure the values are averaged over time, and here? Please make that clear in the text or caption of figures. Please correct also in this caption and next one that the overpass time of OMI is 13.30LT or 1.30pm. Response: Following another reviewer's recommendation, this figure has been removed.

If I understand Figure 7 correctly it can be simplified by basically having the NO2 (ppb) in the x-axis instead of shape factor value. Response: The NO2 shape factor and mixing ratio values provide different information. The two quantities differ by a scaling factor (the total NO2 column). Choice was made to present both of them in the paper.

In Figure 8 it's not clear what values are kept constant and which are changed. Is it the case that the reference has: SSA=0.91, SZA=40âŮę, and surface reflectance=0.03? This info should be more clear in the text (specially) or figure. Response: This has been clarified in the revised manuscript in Section 6:"The reference solar zenith angle (SZA), SSA and surface reflectance are 40°, 0.91, and 0.03 respectively. In the sensitivity experiment, each parameter varies while others are kept constant."

3) Although the chemical compounds are normally known to the scientific community it is good practice to write the name such as for HOx, PAN, HCN, etc. The same for used acronyms or variables such as: Slt, OMI DP GC, TM4 (P6659, L5, L22, L23, respectively), MODIS (P6660, L2), GOES (P6662, L26) Response: Full names have been added for MODIS, GOES, Slt (DP GC, TM4 do not appear anymore in the revised manuscript).

4) Typos to be corrected across the paper such as, aircraft (twice at least), understanding (P6650, L11). Response: These have been corrected throughout the manuscript.

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5) Throughout the paper often I came across incomplete information such as: P6646, L11: which emission inventory is that?; P6658 L5, what NO2 climatologies? (at least a reference should be mentioned). Response: The inventory used for biomass burning emissions (FLAMBE) is now explicitly named in the abstract. Also, in Section 9 the NO2 shape profiles climatology used is specified along with the corresponding reference (Martin et al., 2003).

6) I believe it is more correct and complete to say “solar backscatter radiation”, “extinction/scattering coefficient”, “aerosol correction factor”. Response: These have been corrected.

7) P6646, L2: NO2 AMF depends on more than just the 2 mentioned issues so for completeness, add “among others” or “for example” (same for P6647, L14) Response: It is now said:" The accuracy of space-based nitrogen dioxide (NO2) retrievals from solar backscatter radiances critically depends on a priori knowledge of the vertical profiles of NO2 and aerosol optical properties."

8) P6646, L20-21: rephrase as it looks like aerosol itself is sensitive to surface reflectance Response: The sentence is now:"The effects of aerosol and shape factor are most sensitive to surface reflectance and clouds."

9) P6647, L5: replace “space and time variability” by “spatial and temporal variability”. Response: This sentence does not appear anymore in the revised introduction.

10) P6647, L20: several CTMs already represent fire events, therefore rephrase as “not resolved” is not fully true. Response: This has been nuanced in the revised introduction.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/6/C4291/2014/amtd-6-C4291-2014-supplement.pdf

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