Interactive comment on "Impact of aerosols on the OMI tropospheric NO_2 retrievals over industrialized regions: how accurate is the aerosol correction of cloud-free scenes via a simple cloud model?" by J. Chimot *et al.*

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We thank Referee #2 for his or her comments and suggestions. Below we address them one by one (Referee # 2 comments in blue, author and co-authors answers in black).

General remarks:
This paper presents useful results on a critical aspect of NO_2 retrievals over polluted areas: the impact of aerosols and its implicit inclusion through the modified retrieval of cloud parameters. In particular, the authors quantify the error associated with the implicit correction by comparing the latter with an explicit aerosol correction. This is obtained by using an explicit radiative transfer calculation of the aerosol optical effects as input to the OMI cloud algorithm. While relevant for the community, this paper is not suitable for publication in AMT in its current form. I believe the paper will be publishable in AMT after the authors address the following recommendations. The English should be carefully checked throughout the paper, preferably by a native speaker. There a many typos. Surprisingly, the authors do not cite some recent studies focusing on the effect of aerosol on NO_2 retrievals and therefore fail to provide the necessary discussion comparing their results with previous ones. In particular, the authors should clearly differentiate between new results and confirmation of previous findings. Some sections are very dense and confusing for the reader and need to be more synthetic.

We have carefully checked the English throughout the paper and corrected the typos when found.

With respect to the comments about the citation of recent studies, we would like to emphasize that several recent and relevant studies, focusing on the same problems, have been cited in our initial version of the paper. In particular:

- Castellanos *et al.*, (2015) who studied the effects of absorbing aerosols on the OMI tropospheric NO_2 retrievals over South-America;
- Lin *et al.*, (2014) and Lin *et al.*, (2015) who evaluated the changes of OMI tropospheric NO_2 columns over China by considering explicitly aerosol effects from different dataset
- Kuhlmann *et al.*, (2015) who recalculated tropospheric NO_2 AMFs using high-resolution aerosol parameters over the Pearl River Delta region in southern China;
- Wang *et al.*, (2015b) who analysed aerosol and NO_2 MAX-DOAS data over Wuxi city area with high pollution adjoined to Shangai;
- Shaiganfar *et al.*, (2011) and Kanaya *et al.*, (2013) who identified negative biases of around between -26 and -50% on the OMI tropospheric NO_2 vertical columns in regions with high aerosol pollution ;
- Ma *et al.*, (2013) who explained that these underestimations can be caused by the presence of elevated aerosol layers;
- Boersma *et al.*, (2011) who observed some perturbations of the OMI O_2-O_2 cloud products in presence of aerosols over USA.

These studies were cited in the introduction. They were also inter-compared with our main findings in section 4.1.1., highlighting consistent numbers and conclusions with our sensitivity study. In addition, the study [Leitao *et al.*, 2010] was cited in the introduction and in section 2.2, for explaining the definitions of shielding and enhancement of aerosol particles on the atmospheric properties. This study investigated the explicit impacts of aerosols on the computations of NO_2 AMF as a function of different parameters. Our findings are consistent and confirm the primary importance of the shape of the NO_2 profile and the relative altitude of aerosols to the tropospheric NO_2 bulk as initially pointed out by [Leitao *et al.*, 2010]. We added this comparison in section 2.3.2. to highlight this as recommended by Referee #2.
We think our results are complementary in the sense that our evaluations of the explicit aerosols effects on AMF as a function of AOT are directly compared to the observed implicit effects in the DOMINOv2 product for 2 typical seasons (winter and summer). Moreover, the impact of the monthly variabilities of the NO_2 profile shapes, as given by the TM5 Global Atmospheric Transport Model, on the AMF has been investigated and added on the different figures. This part is new compared to the studies mentioned by Referee #2.

We have now added [Bousserez, 2014] in the introduction, where we listed some recent studies focusing on the problems of aerosols and tropospheric NO_2 retrievals and in the first section.

Please note that Referee #2 refers to Bousserez, (2015) in all the comments below. However, we never found that reference online except one that was focused on CH_4 observations. We assumed that Referee #2 instead wanted to mention the study Bousserez, (2014) "Space-based retrieval NO_2 over biomass burning regions: quantifying and reducing uncertainties". This is the only reference we found from this author focusing on tropospheric NO_2 retrieval.

More details are given below:
Detailed comments:
- The citation format inside a sentence is Author et al., (date), not (Author et al., date). There are many instances of this error in the manuscript.
This is now corrected in our new version.

- Page 8388, line 22: a least one sentence explaining the impact of aerosols on the NO_2 retrievals is needed before the last sentence of the paragraph.
We moved this last sentence to the next paragraph, after the sentence "Aerosols affect the top-of-atmosphere radiances in the visible and O_2-O_2 spectral bands".

- Page 8389, line 17: Bousserez (2015) focused on the impact of aerosols on NO_2 retrievals and should be cited here.
We added this.

- Page 8389, line 23 to end of section: Please provide a number for each section.
Done.

- Page 8390, line 8-9: Those two sentences are not needed.
They are removed.

- Page 8390, line 22: replace "geophysical conditions" with "optical properties of the atmosphere ".
We added the following "accurate knowledge of all the parameters affecting the optical properties of the atmosphere and then impacting the length of average light path".

- Page 8391, line 10: Figure 2 appears in the text before Figure 1. Please correct the order of the figure.
Done

- Page 8391, line 11-12: Not, from Fig. 2, the scattering weight (a(p)) values decrease toward the surface with or without the presence of aerosols. Also, it is odd to comment on Fig. 2 at this point since this section is dedicated to the description of the algorithm to compute the AMF.

The decrease of a(p) toward the surface is observed for a clear scene (without aerosols) and also with aerosols. We added this comment. We moved this comment to the end of section 2.2.

- Page 8391, line 15 to end of section: this discussion is a bit confusing. The satellite measurement is sensitive to both the NO_2 shape profile and the scattering weights. I do not understand why the authors try to separate those effects here and linger on this discussion.

As described in [Wagner *et al.*, 2007; Rozanov *et al.*, 2010; Richter and Wagner, 2011], the AMF (named here A) is obtained from the box-AMF, or altitude-resolved AMF for an atmospheric layer (named here a(p)). It is deduced from the product of the box-AMF by the vertical profile of the considered trace gas. The altitude-resolved air mass factor gives the changes in the slant column density for a change in the vertical column density at the atmospheric layer p, as described in equation 2. We believe that by "scattering weights", Referee #2 means the so-called weighting functions [Richter and Wagner, 2011]. These functions are used in some retrieval algorithms (like the Optimal Estimation Method) which are more adapted for retrieval of strongly absorbing trace gases like O_3 (but not NO_2). The weighting functions correspond to the derivative of the spectral intensity as a function of change of the vertical column density at an atmospheric layer p. As mentioned by [Richter and Wagner, 2011], it is closely related to the Box-AMF in that it only describes changes in intensity, and not in slant column density. The concept of altitude-resolved AMF is well known and acknowledged by the whole DOAS retrieval community [Platt and Stutz, 2008], and includes so by definition the impact of weighting functions which vary for geometry angles, surface albedo, temperature, pressure, presence of particles.

The ratio of the altitude-resolved air mass factor to the total AMF (deduced from the NO_2 shape profile or partial sub-columns) gives the averaging kernel vector, *i.e.* the sensitivity of the satellite measurement to each atmospheric layer [Eskes and Boersma, 2003]. Therefore, as written in line 15, assuming constant NO_2 profile, the altitude-resolved gives an indication of the sensitivity of the satellite measurement to the amount of NO_2 at all the atmospheric levels (and so in the lowest atmospheric layers) [Richter and Wagner, 2011].

We think that our description of AMF and altitude-resolved AMF well reflects this explanation supported by the mentioned references. We added, for a better readability, an explanation about the link between AMF, Box-AMF and averaging kernel as supported by [Eskes and Boersma, 2003]. The concept of weighting functions is not added in our section, to avoid confusion for a general reader, since this concept is not directly used in a DOAS retrieval approach (like in the DOMINO v2).

- Page 8392, line 19: "in the OPPOSITE direction".

In presence of aerosol or cloud particles, photons coming from the top of the atmosphere are usually scattered back towards the satellite, while those reflected from the surface are scattered back towards the surface.

We modified "in the direction of the satellite" accordingly to this.

- Page 8394, line 8-16: please summarize the description of the set up in a table. Also, it would be useful to give more details on the model simulation in an Appendix.

Details about the model simulation are given in 2.3.1. For more details about the DISAMAR model, we encouraged the reader to read [de Haan, 2011; de Haan *et al.*, 1987; Stammes *et al.*, 2001] as referred at the beginning of the section.

Furthermore, for each specific figure, (and associated analyses), we wrote all the specified parameters either in the captions or in some legends. Please note that some parameters differ from one figure to another. We find that repeating all these parameters in another (or new) table would bring no new information and would make this paper too heavy. If nevertheless some essential details are still missing, we would be happy to add them where appropriate.

- Page 8394, line 23 to end of section: This paragraph should belong to the next "Results" section.
This is moved to the next section.

- Page 8395, "Results" section: the authors should cite Leitao et al., (2010) and Bousserez (2015) here, and compare their results with those studies. In particular, results described in line 12-25 are not new.
As notified previously, we added the studies Leitao *et al.*, (2010) and Bousserez, (2014) (we believe here that Referee #2 meant 2014, not 2015). Our results are very consistent with the findings of [Leitao *et al.*, 2010]. Again, these results are complementary in the sense that 2 typical NO_2 profiles of each season (1 in July, 1 in January) are considered in order to evaluate the differences induced by the shape of NO_2 profiles. Bousserez, (2014) did a small exercise assuming one aerosol profile (and so only one or 2 AOT values, not found in the paper) and one NO_2 profile.
Moreover, our present study makes use of monthly variabilities of the NO_2 profiles, as given by TM5, in addition to variable AOT and aerosol altitude which were not considered in Bousserez, (2014). These evaluations can then be directly compared to what seen in the DOMINOv2 product, as done in section 3.

- Page 8396, line 1-15: The conclusions presented here seem similar to what was found in Bousserez (2015). Please discuss that.
The conclusions here are in line with those of Bousserez, (2014) as depicted in Figure 4. However, our study cases include more AOT and aerosol altitude values (compared to 1 single elevated aerosol layer as illustrated in Figure 3 in Bousserez, (2014)), and different NO_2 vertical profiles. This allows then to see the impact of surface albedo, and aerosol properties in case of shielding and enhancement effects. Furthermore, the impact of particle size, through the specification of the Angstrom coefficient, is not present in Bousserez, (2014).

- Page 8396, line 14-15: Not sure what this means. Please rephrase.
By looking at figures 3 and 4, we can see the variability of f for a given AOT and aerosol altitude (expressed through the error bars). As explained at the end of section 2.3.1., this variability is directly linked to the variability of the TM5 NO_2 profiles over 1 month.

- Page 8397, line 19-20: What figure are you referring to? Figure 6? This is not mentioned.
We refer to Figure 6. This is added.

- Page 8397, line 25: it seems "tropospheric NO_2" should be replace by "AMF" here.
We replaced 'tropospheric NO_2 A' by 'A'.

- Page 8398, line 3-7: Explain why.
Here, we show that the retrieved effective cloud fraction values are higher over dark surfaces (i.e. low surface albedo) than over bright surfaces. This is a direct consequence of the aerosol effects on the continuum reflectance. The surface reflectance is more attenuated when aerosol particles are over a bright surface and therefore, the associated effective cloud fraction is reduced. On the contrary, over dark surface, the scattering effects of aerosols increase the fraction of photons that are scattered back towards the satellite sensor. Therefore, in that case, aerosols play the role of a surface with a higher albedo as they increase the brightness of the scene. As a consequence, the retrieved effective cloud fraction value is higher.
All these effects are demonstrated and explained in the section 3.3.1 (Response of the cloud fraction to aerosol scenes) and our analysed study cases.

- Page 8398, line 9-10: Which figure are you commenting?
We are referring to Figs 8 and 9. They are added.

- Page 8398, line 15-20: A bit confusing and unecessary. We already know that the cloud retrieval is sensitive to aerosols. Also, the shielding effect does not depend on the season but rather on both the aerosol and NO_2 profiles (which may have different characteristics depending on the season).
In this section, we analysed the behaviour of the effective cloud parameters, as present in the DOMINOv2 product, in presence of aerosol particles. Indeed, previous papers already mentioned that these parameters are perturbed by aerosols, but the way they behave, depending on the surface albedo and/or aerosol properties, was not explained or analysed in depth. We did this here, and we wanted to point out, in summary, that the implicit aerosol correction, based on perturbed cloud retrievals, statistically applies a shielding effect. Moreover, we have shown that the behaviour and magnitude of this shielding effect with increasing AOT is not similar between summer and winter over China. This is a consequence of aerosol and NO_2 vertical profiles which vary depending on season. As a consequence, the implicit aerosol correction currently applies an increasing shielding effect with increasing AOT values in summer, while it statistically stays constant in winter.
We think these explanations add new information compared to our current knowledge on the behaviour of the effective cloud parameters over cloud-free scenes, dominated by aerosol particles.

- Page 8399, line 25-28: Not clear, please rephrase.
We reformulated as follows: the effective cloud pressure is mainly constrained by the perturbation of the clouds on the O_2-O_2 collision complex absorption. A cloud located at high altitude shields the O_2-O_2 complexes that are below the cloud. As a consequence, the O_2-O_2 absorption signal, and so the associated slant column density, are attenuated.

- Page 8401, section 3.3.1: Is the right surface albedo used for the cloud retrieval in those experiments? In other word are all parameters identical in the aerosol effects and cloud parameters computations?
Yes. All the parameters (including surface albedo) are identical in the simulated spectral aerosol effects and the retrieval of effective cloud parameters. Thus, we ensured that the retrieved values are a direct consequence of aerosols effects on the O_2-O_2 continuum reflectance, O_2-O_2 slant column density and the current employed OMI cloud Look-Up Table (LUT).
This precision is added in the section 3.3.

- Page 8401, line 14-16: Explain why.
We already added the corresponding explanation in the previous section (see remarks about Page 8398, line 9-10).

- Page 8402, line 20: Not sure "LUT" was defined before.
No indeed. This is defined now at this line / section.

- Page 8402 to end of section 3.3.2: This part is difficult to read and needs to be more synthetic.
We tried to be more concise in this section. Please see the new version of our paper.

- Page 8405, line 6-13: Those results seem in contradiction with findings in Bousserez (2015) (see Fig. 6), where the shielding effect was shown to enhance the NO_2 shape factor error. Please comment on that.
We do not see a direct link between our findings in our lines and the Fig. 6 in Bousserez, (2014). Figure 6 of Bousserez, (2014) shows the sensitivity of the normalised tropospheric NO_2 AMF (named in our paper factor f) assuming that a cloud is present above scattering aerosols. In our paper, we assume that an opaque Lambertian cloud, with an albedo of 0.8, was considered here (this information is not present in this paper). It is shown that this factor f decreases with increasing effective cloud fraction. In our analyses, we did not consider a cloudy scene mixed with scattering aerosols. We assumed an aerosol-free scene (*i.e.* no aerosols, only an opaque Lambertian cloud as described in lines 17-20 on Page 8404). This also shows that if this cloud is located above the tropospheric NO_2 bulk, the AMF decreases with increasing cloud fraction, while it increases in the case of mixed cloud with NO_2.
On Page 8405, line 6-13, we discussed the impact of the monthly variability of the NO_2 profile (as given by TM5) on the variability of the factor f. The variability of f is therefore higher when clouds are below or

mixed with the tropospheric NO_2 bulk and almost negligible when the cloud is located above (as a result of shielding effect). This effect is to our knowledge not described in Bousserez, (2014).
Thus we do not think there is any contradiction between both studies.

- Page 8405, line 16 and 21: What does "statistic decrease" mean? Please rephrase.
By "statistic decrease", we mean that, on average, the tropospheric NO_2 AMF decreases in summer with increasing AOT. We reformulated this.

- Section 4.1.1: This section is very dense and needs to be more concise.
We tried to be more concise in the section. Please see the new version of our paper.

- Page 8406, line 3-5: Not clear. Please Rephrase.
We reformulated as follows: The implicit aerosol correction on the tropospheric NO_2 retrieval is based on the properties of the OMI inverse cloud model (*i.e.* opaque Lambertian cloud, with an albedo of 0.8) and the retrieved effective cloud parameters. Its accuracy is evaluated and discussed through the computation of the relative bias Sa.

- Page 8406, line 10-12: Check the English.
We reformulated as follows: Then, the implicit aerosol correction can be compared to the case of no aerosol correction.

- Page 8406, line 20 to end of paragraph: Again, these facts are discussed in Leitao *et al.* (2010) and Bousserez (2015). Please cite and compare when appropriate.
This section discusses the bias induced by the implicit aerosol correction as implemented in the operational OMI NO_2 retrieval chain (through the use of retrieved effective cloud parameters over scenes dominated by aerosols). Both Leitao *et al.*, (2010) and Bousserez, (2014) studies did not address the differences between the explicit and implicit aerosol corrections over non-cloudy scenes but focused on the explicit aerosol effects on the computation of the tropospheric NO_2 AMF. At the end of their study, Leitao *et al.*, (2010) recommended the following in the conclusion section: "The presence of aerosols will also impact on the retrieved cloud properties which in part can compensate the aerosol effects in the absence of real clouds. The details of the interplay between aerosol effects and cloud correction algorithms are complex and should be investigated in more detail". Our study thus followed this recommendation. In section 7, Bousserez, (2014) discussed the potential impact of the presence of cloud above a scattering aerosol without giving any demonstration. But in our study, we did not investigate cloudy scenes, but only cloud-free scenes with presence of aerosols.
Therefore, we do not think that it is appropriate to cite these studies in that section.

- Page 8409, line 1: the authors should also mentioned results obtained in Bousserez (2015) above biomass burning here.
Again, in that section, we are analyzing the implicit aerosol correction applied on the OMI tropospheric NO_2 retrievals, through the effective cloud parameters perturbed by the presence of aerosols. This is compared to an explicit aerosol correction (*i.e.* using explicitly aerosol parameters) and no aerosol correction. As far as we could see, Bousserez, (2014) did not explicitly address the problem of implicit aerosol correction and the link between the aerosols and the retrieval of effective cloud parameters contrary to the other mentioned studies which reprocessed DOMINO dataset by applying explicit aerosol effects. A few assumptions are given in section 7 but not demonstrated. Moreover, this section is more focused on the presence of clouds in addition of aerosol particles. This is not the case here. The [Bousserez, 2014] study focused on NO_2 retrieval errors because of misrepresentation of the NO_2 shape profile. Thus, we do not think it is relevant to cite this study here on top of our citation in the introduction.

- Page 8409, line 17: What does "irregular behavior" mean? Figures and Tables:

By “irregular behaviour”, we mean that the relative biases associated with the implicit aerosol correction do not smoothly increase or decrease with increasing AOT. They somewhat either increase or decrease depending on AOT (and aerosol altitude) values. As written in the next sentences, this differs from the relative biases associated to cases with no aerosol correction that smoothly increase with increasing AOT.

- Table 1: Replace "requested" by "required" in the legend. Please do not use qualitative statements in the table, but rather quantify the uncertainties for each parameters.
We replaced “requested” by “required”.
The idea of this table is double: 1) to list most (if not all) of the parameters that are necessary for the computation of the altitude-resolved air mass factor (*i.e.* a(p)), 2) to classify the degree of certainty of these parameters. Such a degree allows then, for a general reader, to deduce which of these parameters currently contribute to the highest uncertainty when computing the tropospheric NO_2 AMF. Based on our study and all the mentioned reference papers, it is today well known that aerosols and NO_2 profile are the most uncertain parameters. The impacts of aerosols (and somewhat clouds) are discussed quantitatively in this paper. The global uncertainty of the DOMINOv2 product is also discussed in the introduction section.
To give quantitative uncertainties of each of the other parameters is clearly out of the scope of this present study. This would require to inter-compare a lot of studies done since the last years, on different measurements and using different approaches, and this is not the purpose of this table devoted to a general reader.

- Modify order of Figure 1 and 2, which do not appear in this order in the text.
Done

- Figure 2: Use "Pressure" instead of "Vertical pressure profile" for the Y-axis.
Done

- Figure 3: "...as a function of AOT AND AEROSOL LAYER ALTITUDE...
Added

Proposed additional references

Eskes, H. J., and K. F. Boersma (2003), Averaging kernels for DOAS total column satellite retrievals, Atmos. Chem. Phys., 3, 1285–1291.

Richter A. And Wagner T., Chapter 2 “The Use of UV, Visible and Near IR Solar Back Scattered Radiation to Determine Trace Gases”, extracted from J.P. Burrows et al. (eds.), The Remote Sensing of Tropospheric Composition from Space, Physics of Earth and Space Environments, DOI 10.1007/978-3-642-14791-3_2, Springer-Verlag Berlin Heidelberg 2011.