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 #1 for his or her valuable comments and suggestions. Below we address them one by one (Referee # 1 comments in blue, Author and co-authors answers in black).

The paper analyzes the complex effects of aerosols on cloud and NO_2 retrievals from OMI, by doing sensitivity tests with assumed aerosol and atmospheric properties. It can be published after some revisions and clarifications.

The sensitivity tests follow the DOMINO procedure, especially the use of a LUT. And the authors point out the significant limitation in the LUT that undermines the analysis of the actual effects of aerosols on cloud and NO_2 retrievals. While analyzing the behavior of DOMINO (with its LUT) is interesting, a general reader can benefit from additional sensitivity studies with no use of the LUT. In this way, a more general question can be answered on how aerosols affect cloud and NO_2 retrievals (rather than how DOMINO is limited by its aerosol treatment with its particular LUT). Also, since this LUT limitation is important, it appears appropriate to indicate such limitation in the abstract.

The understanding of Referee #1 is fully correct. One of the key messages pointed out by our study is the limitation of the employed OMI cloud LUT when retrieving tropospheric NO_2 columns over clear scenes dominated by aerosol particles. We agree with Referee #1 that this message should be included in our abstract. It is added now.

The retrieval of effective cloud parameters generally requires the use of a LUT for being able to convert the continuum reflectance and the O_2-O_2 Slant Column Density (SCD) into effective cloud fraction and pressure values. Since different LUTs may give different results, we have done the following exercise as illustrated in Figure 1 and Figure 2 in order to illustrate in a general way how aerosols affect the OMI effective cloud retrievals,:

- ✓ We simulated spectra in the O_2-O_2 spectral band (from 460 nm to 490 nm) containing an opaque Lambertian Cloud (albedo = 0.8) assuming different cloud fraction and cloud pressure. The considered values are described in Table 1. A high sampling in cloud fraction values was considered for these simulations. Aerosols are not present in these simulations.
- ✓ Similarly, we simulated cloud-free spectra dominated by aerosols assuming fine particles (Angstrom Coefficient = 1.5), and high scattering properties (Single Scattering Albedo = 0.95). Different Aerosol Optical Thickness (AOT) and Aerosol Effective Height (AEH) are considered (see Table 1). Aerosols are considered to be present in box atmospheric layers. A high sampling in AOT values is here considered.
- ✓ A DOAS fit is achieved for all the simulations in order to derive the continuum reflectance at the reference wavelength (475 nm) and the O_2-O_2 Slant Column Density (SCD).

- ✓ Then, effective cloud fraction, effective cloud pressure, AOT and AEH values are linearly interpolated / extrapolated in order to have a global overview of the variation of these variables as a function of the DOAS fit variables.

Figure 1 illustrates that the effective cloud fraction value is primarily constrained by the continuum reflectance, while the O_2-O_2 SCD mainly drives the effective cloud pressure magnitude. Similarly, AOT value mostly impacts the continuum reflectance magnitude while the aerosols altitude (or AEH) mostly results in a change of O_2-O_2 SCD value (*cf.* Figure 2). Furthermore, in the case of low continuum reflectance (below than 0.2), which corresponds to aerosol cases, and low effective cloud fraction, it can be observed that there are some correlations between the derived O_2-O_2 SCD and continuum reflectance retrievals.
Therefore, in the case of an ideal O_2-O_2 cloud retrieval (*i.e.* without the specific limitation f the analyzed LUT), the following is expected:

- ✓ For a given aerosol altitude value, increasing AOT should result in a larger continuum reflectance and thus increases the effective cloud fraction value;
- ✓ For a given AOT value, increasing the aerosols altitude (or decreasing AEH) should result in smaller O_2-O_2 SCD and therefore decreases the effective cloud pressure value;
- ✓ Since increasing AOT primarily impacts the continuum reflectance but also simultaneously impacts the O_2-O_2 SCD value, retrieved effective cloud pressure theoretically could either increase, decrease or stays constant depending on the aerosols altitude: this demonstrates that the magnitude of the O_2-O_2 shielding by aerosols is a combination of aerosol amount and altitude.

The high sampling of simulations as depicted in Figure 1 and Figure 2 shows that the variation of effective cloud pressure in case of low continuum reflectance has very small impacts on the O_2-O_2 SCD. However, it is still theoretically possible to retrieve small values (not only values close to the surface). Low sampling of simulations results in inaccuracy of the interpolation / extrapolation between the simulation nodes. This is why the current OMI cloud LUT exhibits higher effective cloud pressure values.

We propose to add these figures and the associated analyses, written just above, in our paper just after the sub-section 3.3.2. This will be a new sub-section: 3.3.3: Comparison of cloud and aerosol impacts on the O_2-O_2 spectral band. We think then that this new subsection can describe to a general reader how aerosols are expected to impact the cloud retrievals without the current limitations of the LUT.

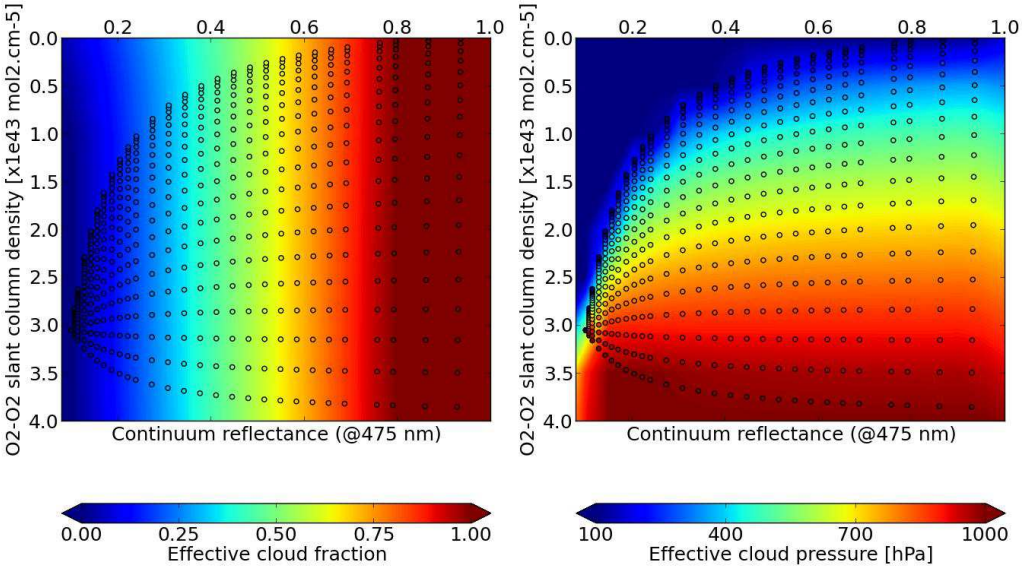

Figure 1: Effective cloud fraction and cloud pressure as a function of O_2-O_2 slant column density and continuum reflectance assuming an opaque (albedo = 0.8) Lambertian cloud model and the following conditions: temperature, NO_2, O_3 and H_2O profiles from US standard 1976, surface albedo = 0.05, SZA = 32 deg, VZA = 32 deg, altitude = 0 km. The dots represent the values specified in the forward simulations (named simulation nodes). The background colors are the results of the linear interpolation / extrapolation of the DOAS fit results shown.

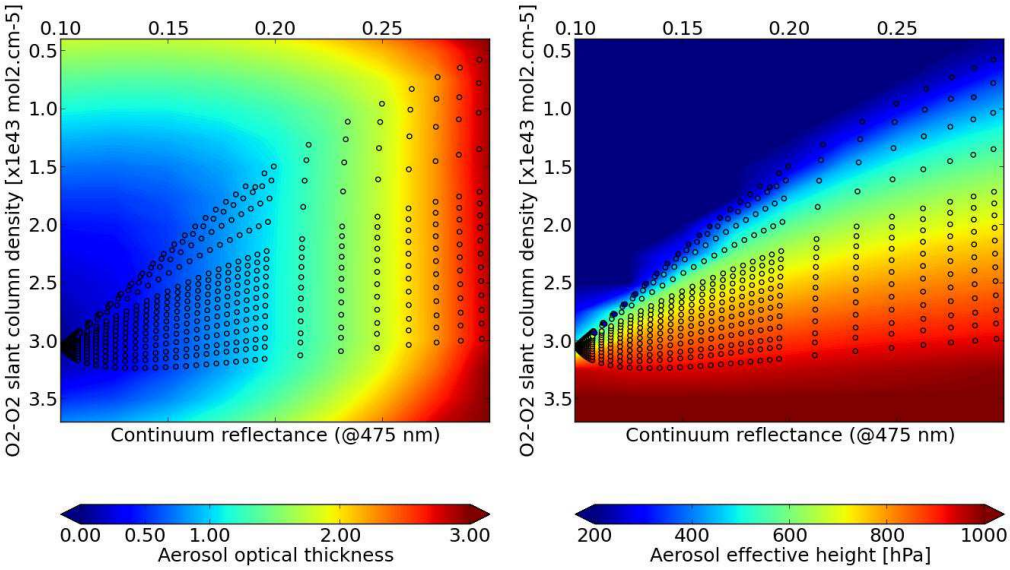

Figure 2: Aerosol optical thickness and effective height as a function of O_2-O_2 slant column density and continuum reflectance assuming fine and very scattering particles (SSA = 0.95, Angstrom coefficient = 1.5, asymmetry parameter = 0.7) and the following conditions: temperature, NO_2, O_3 and H_2O profiles from US standard 1976, surface albedo = 0.05, SZA = 32 deg, VZA = 32 deg, altitude = 0 km. The dots represent the values specified in the forward simulations (named simulation nodes). The background colors are the results of the linear interpolation / extrapolation of the DOAS fit results shown.

Parameter	List of values
Effective cloud fraction	0., 0.01, 0.02, 0.04, 0.06, 0.08, 0.1, 0.125, 0.15, 0.175, 0.2, 0.25, 0.3, 0.35, 0.4, 0.45, 0.5, 0.55, 0.6, 0.65, 0.7, 0.75, 0.8, 0.95, 1., 1.1, 1.2
Effective cloud pressure [hPa]	1013, 963, 913, 863, 813, 763, 713, 663, 613, 563, 513, 463, 413, 363, 313, 263, 213, 163, 113, 63
Aerosol optical thickness	0., 0.01, 0.02, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08, 0.09, 0.1, 0.15, 0.2, 0.25, 0.3, 0.35, 0.4, 0.45, 0.5, 0.55, 0.6, 0.65, 0.7, 0.75, 0.8, 0.85, 0.9, 0.95, 1., 1.05, 1.25, 1.5, 1.75, 2., 2.25, 2.5, 2.75, 3., 3.25, 3.5, 3.75, 4.
Aerosol effective height [hPa]	975, 950, 925, 900, 875, 850, 825, 800, 775, 750, 725, 700, 675, 650, 550, 450, 350, 250, 150

Table 1: List of values considered for the simulation nodes illustrated in Figure 1 and Figure 2: effective cloud fraction and effective clod pressure for Figure 1, aerosol optical thickness and aerosol effective height for Figure 2.

A few recent studies that addressed the effects of aerosols on cloud and NO_2 retrievals (Lin *et al.*, 2014 for China; Castellanos *et al.*, 2015 for South America; Lin *et al.*, 2015 for China) have been discussed in the present paper (in the end of Sect. 4). It is appropriate to discuss these prior studies in the introduction section, particularly that these works have addressed the effects of aerosols on cloud and NO_2 retrievals. The current writing of introduction is vague and could be read mistakenly as if the present paper is the first study on this topic.

We agree that our study is not the first one addressing the problems induced by aerosols on NO_2 retrievals from satellite UV-Vis measurements. We discussed the main findings of the studies (Lin *et al.*, 2014 for China; Castellanos *et al.*, 2015 for South America; Lin *et al.*, 2015 for China) in sections 3 and 4 and compared with our analysis.

In agreement with the comments of Referee #1, we summarized in the new version of our paper these main findings in the introduction section. But we think that some details should be left in sections 3 and 4 in order to avoid having a too long introduction for the reader. We hope however that the current writing is less vague.
However, although some interplay between aerosols and OMI cloud products have been identified by these cited papers by analyzing real data, at our knowledge our study is the first one analyzing in detail and explicitly how the current operational OMI O_2-O_2 cloud algorithm responses to the presence of aerosols and identifying the main limitations. This follows the recommendations written in the previous cited publications.

As the authors (and previous studies) point out, the relative height of aerosols versus NO_2 is very important when determining whether an implicit aerosol treatment leads to underestimated or overestimated NO_2 VCDs. The work of Vlemmix *et al.* (2015) is often referred to in the present paper to argue that aerosols are above NO_2 in summer in East China. Vlemmix *et al.* (2015) only analyzed MAX-DOAS measurements in Beijing with limitations in observations (MAX-DOAS measurements have difficulties in determining vertical profiles) and location (vertical profiles in Beijing may not fully represent East China). Also, the assumption of aerosol altitude in the present study (*i.e.*, aerosols are evenly mixed within a particular pressure range) differs from the actual vertical profile. Therefore, rather than giving a strong statement regarding NO_2 retrieval bias that has to assume aerosols to be above NO_2, it appears more appropriate to focus on how the relative height would affect the NO_2 retrieval.

Indeed, as we analysed here in this study, the sign and the magnitude of the bias of the retrieved tropospheric NO_2 column depends whether aerosols are located above or below the tropospheric NO_2 bulk and the relative distance between these 2 parameters. Therefore, the relative height of NO_2 *vs.* aerosol particles is the key driver of this bias. It is more important than the absolute altitude of tropospheric NO_2 bulk and aerosols.

At the end of page 8403, we mentioned that retrieved effective cloud pressure values, as present in the OMI DOMINO product, increase with increasing MODIS AOT in summer while they stay close to the surface for all AOT values in winter. According to our sensitivity analyses, effective cloud pressure values are somewhat close to the aerosol altitudes for large AOT values. These data can show then that, in general, aerosol particles are located at higher altitude in summer. This is also a consequence of the lifetime of aerosols (which is longer than NO_2) and the fact that the boundary layer is generally deeper in summer due to convective growth.
We referred to the study of Vlemmix *et al.*, (2015) that shows that in summer in China aerosol particles are generally located higher in altitude than the tropospheric NO_2 layers. It is true that MAX-DOAS data have some difficulties to derive NO_2 and aerosol profiles and that measurement in Beijing may not be fully representative of all East-China.

However, other papers mentioned similar conclusions and we would like to refer to them in addition. Li *et al.*, (2013) performed MAX-DOAS measurements during the PRIDE-PRD2006 campaign in the Pearl River Delta region,in China, for 4 weeks in July 2006. The considered site is located at 60 km north of Guangzhou in a rural area. Figure 6 of this paper clearly shows that (for this data) aerosol mixing layers (grey bars) are the most often deeper / higher than NO_2 mixing layers (blue bars).
Mendolia *et al.*, (2013) retrieved tropospheric NO_2 vertical column densities from OMI and MAX-DOAS measurements over Canada. One key conclusion of this work, as illustrated in Figure 4 c, d, is that NO_2 diurnal profiles can even be systematically lower in summer and do not follow the 'expected' pattern of the convective boundary layer (higher in summer than in winter). Aerosols do follow this seasonal pattern since they have a longer life time.

Abstract
Please discuss the limitation of this study due to use of LUT.

This is added in the abstract.

Line 6-7: POMINO already accounts for explicit aerosols.
Some products, such as POMINO, take now into account the aerosols. They are based on a reprocessing of existing OMI tropospheric NO_2 product and improve the retrievals (in that case with an explicit treatment of surface reflectance and aerosols).
However, here in lines 6-7, we wanted to point out that explicit treatment of aerosol effects are not taken into account in operational tropospheric NO_2 retrievals (such as the DOMINOv2). This is clarified in our abstract.

Line 12: please define 'cloud-free'
We define cloud-free scenes as clear scenes without presence of clouds in the observation pixel. In our study, we focused on scenes with effective cloud fraction values smaller than 0.1 to ensure a high probability to analyze clear OMI scenes, but dominated by aerosol particles.

Line 16: please remove 'linear'. Obviously the relation is not simply linear
This is removed. We wanted to highlight that, assuming uniform aerosol properties, geometry angles, and surface albedo, the achieved experiments have shown an almost linear relation between AOT and retrieved effective cloud fraction. But variability in all these parameters indeed remove this linear relation.

Line 19-21: the reduced cloud pressure is primarily because aerosols are set at higher altitudes than NO_2, rather than due to its 'absorbing effects'
We corrected this. The reduced effective cloud pressure expresses an enhanced shielding effect of the O_2-O_2 column induced by the aerosol particles, not absorbing effects of aerosols. This generally results from a combination of aerosol amount (AOT) and the altitude of aerosols.

Line 21-24: 'actual' here is not clear –you are not doing an actual retrieval. Also, please change 'high aerosol pollution …and elevated particles' to 'high aerosol pollution …at elevated altitudes'
This is removed and corrected.

P8388, Line 9: change '(±25%)'to '±25%'
Done

P8388, Line25 –P8389, Line 19: please update this paragraph to better reflect the existing relevant works on the effects of aerosols on cloud and NO_2 retrievals (Lin *et al.*, 2014; Castellanos *et al.*, 2015; Lin *et al.*,2015). The current writing of introduction is vague and could be read mistakenly as if the present paper is the first study on this topic.
As discussed previously, this part was modified. We hope now that the introduction would not b read mistakenly.

P8390, Line 11: A middle step is to remove stratospheric SCD to derive tropospheric SCD.
Yes this is true. However, in that section, we wanted to address the general discussion about how to convert a NO_2 slant column Density to Vertical Column Density and so focus on the importance of the computation of the AMF. We however added a line indicating this middle step.

Eq. 3 –there is a temperature correction for a(p)
In the case of the OMI tropospheric NO_2 retrievals, the temperature correction is applied on the slant column density, based on the ECMWF temperature fields combined with the a priori NO_2 profiles shapes. This step is necessary as the temperature in the NO_2 absorption cross section is assumed to be fixed at 221 K. A correction term is thus implemented in the computation of A (and not exactly on a(p)) such as it represents the ratio of the NO_2 slant column derived with a NO_2 cross section at T to the column derived at 221 K. This explanation and the description of the correction term is now added [Boersma *et al.*, 2004].

Sect 2.3 –discussion here does not consider the cloud retrieval yet. Please specify this, for better readability.

This is added at the beginning of the first sentence.

Eq.4 – how will the use of this simplified phase function and g affect the analysis?

The Henyey-Greenstein phase function is quite commonly used the DOAS community for retrieving tropospheric NO_2 columns, such as in the study of Vlemmix *et al.*, (2010) or Castellanos *et al.*, (2015), with explicit aerosol corrections.

With an asymmetry parameter of g = 0.7, the Henyey-Greenstein are known to reproduce quite well the Mie functions, as illustrated in Figure 3 by the PhD work of Martin de Graaf. The small differences between then are not expected to impact significantly the accuracy of the tropospehric NO_2 retrieval and effective cloud parameters through the DOAS approach. This is also confirmed by the consistency between the OMI cloud product as extracted from the DOMINO dataset, and our analyses achieved on aerosol synthetic spectra. Moreover, the exercises done with changes in Angstrom coefficient and Single Scattering values do not show significant changes on the computation of the Air Mass Factors (AMF). This is a sign of the stability of the simulations and computations with this function.

In addition, Castellanos *et al.*, (2015), analysed the impact of decreasing from 0.7 to 0.6 the aerosol asymmetry parameter g used in the DISAMAR radiative transfer model. The impacts are less than 5% on the tropospheric NO_2 AMF for AOT < 0.5, and almost negligible for larger AOT values.

Moreover, in a general way, for absorption in the atmosphere, and thus absorption by NO_2, the only quantity that is relevant is the light path distribution, *i.e.* the distribution of distances travelled by photons in the atmosphere before leaving the atmosphere. The absolute radiance at the top of the atmosphere is not important. This light path distribution is not governed by details in the phase function, but by the single scattering albedo a and by the asymmetry parameter g; and of course by the optical thickness. Those two scattering parameters a and g are included in HG scattering, and therefore it can be used for amf calculations [Spada *et al.*, (2006); Wagner *et al.*, (2007); Private communication with Piet Stammes].

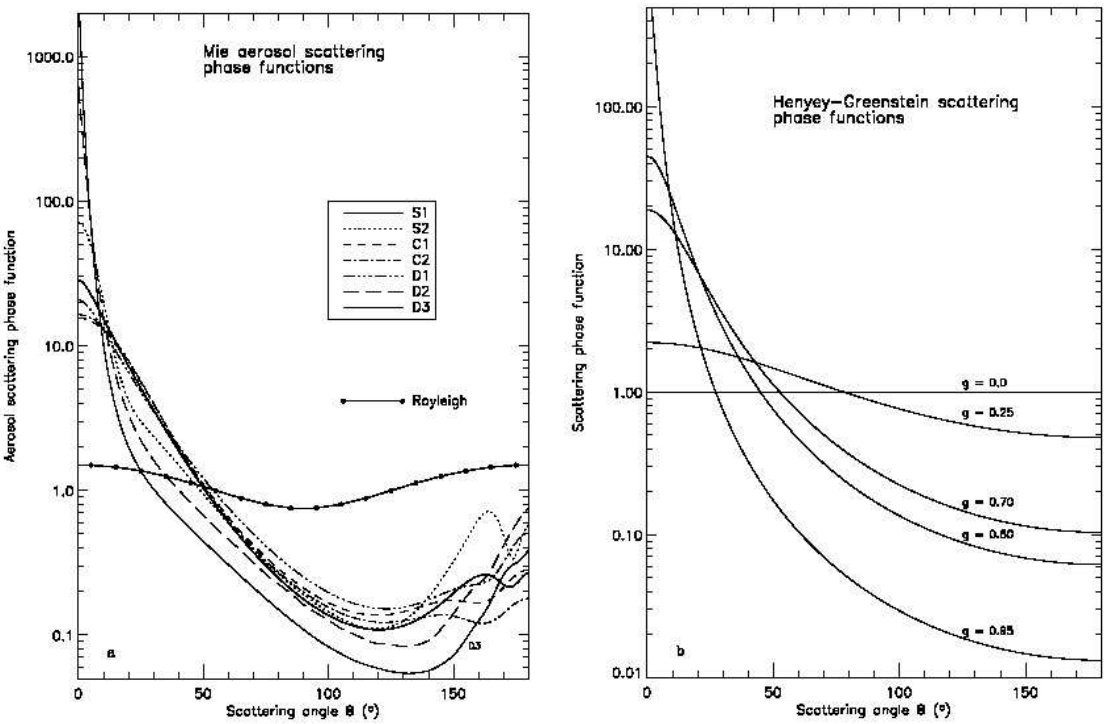

Figure 3: Comparison of aerosol scattering phase function as a function of scattering angle, as simulated with the Mie aerosol scattering phase functions (left) and the Henyey-Greenstein scattering phase functions (right). The Mie aerosol definitions are taken from Torres *et al.*, 1998, JGR 103.

P8394, Line 8 –what is the wavelength for AOD and SSA?
The reference wavelength is 550 nm.

P8397, Line 9 –is this OMI cloud fraction?
The statement following this line indicates that the filter of 0.1 is applied on both effective OMI cloud fraction and geometric MODIS cloud fraction. If one of these parameters indicates a higher value, then both collocated MODIS and OMI observations were filtered out. Note that OMI cloud fraction values are generally higher than MODIS cloud fraction values (in the MODIS L2 aerosol product).

P8397, Line 19-21: any statistical significance? Also, please clarify that here the spatial variability in AOD and NO_2 is included, such that the apparent correlation between AOD and NO_2 may be affected by other spatial factors like albedo, elevation, etc.
The tests done with a higher threshold on cloud fraction (0.2 or 0.3) when selecting collocated OMI and MODIS observations did not show significant statistical differences. At high AOT values, the effective OMI cloud fraction values were somewhat closer to 0.2 or 0.3 (depending on the applied threshold value) but the values were still increasing with increasing AOT. Moreover, similar slopes as described in our paper were observed. Higher effective cloud fraction values at high AOT may also reflect that some cloudy observations were present in the selected observations.
Effective cloud pressure values were statistically very close to the values shown in the figure depicted in our paper.

The clarification asked by Referee #1 is added.

P8398, Line 6: 'loud' should be 'Cloud'
Corrected.

P8401, Line 10: AOT at which wavelength?
AOT values are given for wavelength = 550 nm.

P8402, Line 10-11: how do you know it is due to absorption rather than scattering (since the assumed aerosols are above NO_2)
In this section, we analyse the response of the OMI effective cloud pressure retrieval in presence of aerosols. Such a retrieval is associated with the O_2-O_2 absorption signal and not on the NO_2. The location of aerosols with respect to the tropospheric NO_2 bulk does not have any impact on the cloud pressure retrieval. The behaviour of the OMI effective cloud pressure with increasing AOT is a direct consequence of the shield effect applied by aerosol particles on the O_2-O_2 column.

Since we analyse the effects of scattering aerosol particles, we shouldn't have written "absorption". We think it is more correct to talk about shielding effect by aerosols, since an increasing amount of particles decreases the fraction of photons reaching the lowest part of the atmosphere and increase the attenuation of the surface reflectance signal. We changed this accordingly in this section.

Sect. 3.3.2 –since the analysis is significantly affected by the use of coarse-resolution LUT. Is it possible to do some additional tests with no use of the LUT?
This is done and described in the text above, and in Figure 1 and Figure 2.

P8403, last paragraph and P8405, last paragraph –see my major comments.
Please see or answers above following your major comments.

P8408, Line 8: should be NO_2 AMF
Corrected.

P8409, Line 5-8 –In POMINO, model AOD is constrained by monthly MODIS/Aqua AOD data, and it is also validated by ground-based AOD measurements.
We added this information in our section.

Figure 3 caption –please check the month
The month indicated in the caption of Figure 3 is correct.

Figure 5 caption –should be '0.95 and 0.9'
Corrected

Proposed additional references:

Boersma, K. F., Eskes, H. J., and Brinksma, E. J.: Error analysis for tropospheric NO_2 retrieval from space, J. Geophys. Res., 109, D04311, doi:10.1029/2003JD003962, 2004.

Li, X., Brauers, T., Hofzumahaus, A., Lu, K., Li, Y. P., Shao, M., Wagner, T., and Wahner, A.: MAX-DOAS measurements of NO_2, HCHO and CHOCHO at a rural site in Southern China, Atmos. Chem. Phys., 13, 2133-2151, doi:10.5194/acp-13-2133-2013, 2013.

Mendolia, D., D'Souza, R. J. C., Evans, G. J., and Brook, J.: Comparison of tropospheric NO_2 vertical columns in an urban environment using satellite, multi-axis differential optical absorption spectroscopy, and in situ measurements, Atmos. Meas. Tech., 6, 2907-2924, doi:10.5194/amt-6-2907-2013, 2013.

Vlemmix, T., Piters, A. J. M., Stammes, P., Wang, P., and Levelt, P. F.: Retrieval of tropospheric NO_2 using the MAX-DOAS method combined with relative intensity measurements for aerosol correction, Atmos. Meas. Tech., 3, 1287-1305, doi:10.5194/amt-3-1287-2010, 2010.

Spada, F., Krol, M. C., and Stammes, P.: McSCIA: application of the Equivalence Theorem in a Monte Carlo radiative transfer model for spherical shell atmospheres, Atmos. Chem. Phys., 6, 4823-4842, doi:10.5194/acp-6-4823-2006, 2006.

Wagner, T., Burrows, J. P., Deutschmann, T., Dix, B., von Friedeburg, C., Frieß, U., Hendrick, F., Heue, K.-P., Irie, H., Iwabuchi, H., Kanaya, Y., Keller, J., McLinden, C. A., Oetjen, H., Palazzi, E., Petritoli, A., Platt, U., Postylyakov, O., Pukite, J., Richter, A., van Roozendael, M., Rozanov, A., Rozanov, V., Sinreich, R., Sanghavi, S., and Wittrock, F.: Comparison of box-air-mass-factors and radiances for Multiple-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) geometries calculated from different UV/visible radiative transfer models, Atmos. Chem. Phys., 7, 1809-1833, doi:10.5194/acp-7-1809-2007, 2007.