Authors' Response to Reviewer #2

The article *OMI tropospheric NO_2 air mass factors over South America: effects of biomass burning aerosols* by Castellanos et al. is generally well written and investigates the important effects aerosols can have on the retrieval of tropospheric NO_2 columns. I recommend it to be *accepted subject to minor revisions*.

1 General comments

A large part of this study is an evaluation of the uncertainties of

1. the effects of aerosols on DOMINOv2 AMFs

2. the effects of aerosols on the O_2-O_2 cloud pressure.

I would appreciate if the authors would explicitly compare their results to the previously stated uncertainties (e.g., in Boersma et al., 2004, Acarreta et al., 2004, Stammes et al., 2008).

> Please see Page 2701 Line 9-11: "Approximately 66% of the pixels differ by less than ±0.2 (18%), within the 20% estimated lower limit for the AMF uncertainty for polluted scenes (Boersma et al., 2004).
>
> It would be misleading to compare the ability of the O_2-O_2 effective cloud retrieval to resolve aerosol layer pressures to the reported uncertainty for the retrieval of actual clouds. The O_2-O_2 retrieval was not designed or optimized for application to aerosols, thus the uncertainty metric is applicable for clouds only. In fact, our analysis serves as the first validation that the O_2-O_2 effective cloud retrieval does in fact resolve aerosol layer pressures.

Probably, the result of this comparison will be that previous uncertainty estimates were largely under-estimated in the presence of aerosols.

> On average, changes in the AMF were 10% overall when aerosol effects are included, while the stated lower limit for the uncertainty for the AMF is 20%. However, the probability distribution of the AMF differences is skewed (see Figure 7), and increases with AOD (see Figure 10). Thus, large AMF errors can occur under certain circumstances – AOD > 0.6. As the majority (>71%) of the scenes we analyzed had AOD < 0.6, and the mean difference in AMF is less than 10% for this cohort of pixels, we might in fact say that previous uncertainty estimates are largely accurate in the presence of aerosols.
>
> However, as this study focused on a particular subset of retrievals, namely

those containing fresh biomass burning aerosols, such a generalized statement cannot be supported from the results presented here. Our results do allow us to make recommendations for scene selection in which aerosol effects are adequately accounted for (i.e. within current uncertainty estimates) by the effective cloud parameters and independent pixel approximation. We clearly state these recommendations in the conclusions and Table 1.

2 Specific comments

- 2684/02: Not *OMI* observations are essential, but rather more general *satellite* observations; OMI is only one example.

  Change made.

  Page 1 Line 13: "OMI" was replaced with "Satellite"

- 2684/26: The meaning of the phrase *which was the case for the majority of the pixels considered in our study* is not clear.

  Change made.

  Page 2 Line 6: We added the following after the phrase to clarify: "; 70% had cloud radiance fraction below 30%, and 50% had effective cloud pressure greater than 800 hPa."

- 2686/13: It would help if the authors would explain that f_{eff} could also be called *radiance cloud fraction*.

  Change made.

  Page 3 Line 15: We added the following to clarify: "(or simply radiance cloud fraction)"

- 2688/24: The Schreier et al. paper you cite used GOME-2, not OMI. Or, maybe you mean the earlier study by Schreier et al., which was about the tropics, and actually used OMI?

  Change made. The correct citation is:

  Schreier, S. F., Richter, A., Kaiser, J. W. and Burrows, J. P.: The empirical relationship between satellite-derived tropospheric NO_2 and fire radiative power and possible implications for fire emission rates of NO_x, Atmospheric Chemistry and Physics, 14, 2447-2466, doi:10.5194/acp-14-2447-2014, 2014.

- 2690/08: Shouldn't it be *measured* instead of *simulated* reflectance?

  The manuscript is correct here. Replacing 'simulated' with 'measured' reflectance in this sentence gives: "In Eq. (3), R is the measured reflectance best matching the observed reflectance…", which is meaningless.

- 2690/25: To my understanding, O_2-O_2 number density should be a function of the inverse *pressure*, not the inverse *temperature*. Please explain.

  Starting from Eq. (1) in Acaretta et al. (2004), the O_2-O_2 optical thickness ($\tau_{O_2-O_2}(\lambda)$) is proportional to the square of the molecular oxygen number density (n_{O_2}). This equation can be rewritten as the following assuming hydrostatic equilibrium, and taking the identity that the molecular oxygen number density can be expressed as a function of pressure (p), the volume mixing ratio of oxygen ($[O_2]$), Boltzmann's constant (k_B), and temperature (T):

  $$n_{O_2} = \frac{[O_2]p}{k_B T}$$

  $$\tau_{O_2-O_2}(\lambda) = \int_{\ln(p_{bottom})}^{\ln(p_{top})} \sigma_{O_2-O_2}(\lambda) \frac{RT}{gM} \left(\frac{[O_2]p}{k_B T}\right)^2 dln(p)$$

  where g is gravitational acceleration, M is the molecular mass of dry air, and $\sigma_{O_2-O_2}(\lambda)$ is the wavelength dependent absorption cross section of O_2-O_2.

  The above equation shows the inverse temperature dependence of the O_2-O_2 optical thickness, and thus absorption.

  A full explanation of the O_2-O_2 slant column temperature related error can be found in de Haan (2010), but is in the author's opinion outside the scope of the manuscript.

  de Haan, J., Cloud pressure retrieval using O2-O2 at 477 nm, temperature effects, KNMI Technical Document, TN-OMI-KNMI-972, 2010.

- 2691/26: Maybe mention the correction for the temperature-dependence of NO_2 absorption in this context?

  The manuscript includes in the following paragraph an explanation for the correction of the temperature-dependence of NO_2 absorption. Please see Page 2692 Line 11 and Eqs. (4-6) in the manuscript.

- 2692/14: Which ECMWF data? Forecast? ERA-Interim?

Change made.

Page 8, Line 19: “ECMWF operational medium-range forecast data”

- 2692/19: Which wavelength are the NO_2 AMFs calculated for? What influence does the wavelength disparity between the O_2-O_2 cloud retrieval and the NO_2 retrieval have?

  The NO_2 AMF is calculated at 439 nm. Please see Page 2699, Line 19 in the manuscript.

  The O_2-O_2 cloud retrieval considers O_2-O_2 absorption at 475 nm.

  The premise of the O_2-O_2 cloud retrieval and aerosol (as well as cloud) correction in the NO_2 retrieval is that the radiative effects of aerosols at 475 nm can be represented by an opaque Lambertian surface at a certain height covering a fraction of the surface pixel area. If the scattering and absorption properties of the aerosols have a strong spectral dependence, then the effective cloud fraction and pressure that best simulate the vertical sensitivity to NO_2 at 439 nm could be under/over estimated, depending on the nature of the aerosols. However, from our results, we show that for biomass burning aerosols generally this is not the case, as the majority of our results show small (< 20%) differences between AMFs calculated with effective clouds versus observed aerosol parameters.

- 2692/23: Rephrase; on first sight, it looks like the *surface observations* are distinct from the *MAX-DOAS observations*, even though they are the same.

  Change made.

  Page 9, Line 1: “Irie et al. (2012) and Ma et al. (2013) have shown that that DOMINO v2 NO_2 tropospheric columns are highly correlated with the surface MAX-DOAS observations…”

- 2693/07: Please explain what *converted values* are.

  Change made.

  Page 9 Line 9: “AOD at 354 and 500 nm converted from 388 nm are also reported…”

- 2693/20: Which RTM was used?

  Change made.

  Page 9 Line 23: “The algorithm uses a LUT of reflectances at 354 and 388 nm that were calculated for each aerosol model using the University of

Arizona radiative transfer model (Caudill et al., 1997)."

- 2693/26: What does an AI larger than 0.5 mean physically?

  The UVAI is a measure of the deviation of the observed UV spectral contrast from a pure Rayleigh scattering atmosphere. UVAI will be negative for scattering aerosols, positive for absorbing aerosols, and will increase with the height, the optical depth and the single scattering co-albedo of the absorbing aerosol layer (de Graaf, 2005; Torres et al., 1998).

  UVAI greater than 0.5 was chosen as a threshold to indicate elevated aerosols in the OMAERUV retrieval. However, the level 2 data product also provides the AOD and SSA solutions at the five ALH nodal points (0, 1.5, 3.0, 6.0, and 10 km) of the LUT. Thus, one can interpolate the AOD and SSA to an ALH other than the OMAERUV reported best guess ALH if better information on the ALH is available (please see Page 2694 Line 5-10 in the manuscript), which is in fact what we have done in our study.

  M de Graaf, P Stammes, O Torres, and R B A Koelemeijer, Absorbing Aerosol Index: Sensitivity analysis, application to GOME and comparison with TOMS, Journal of Geophysical Research, 110 (D1), D01201, 10.1029/2004JD005178, 2005.

  Torres, O., Bhartia, P. K., Herman, J. R., Ahmad, Z., and Gleason, J.: Derivation of aerosol properties from satellite measurements of backscattered ultraviolet radiation: theoretical basis, J. Geophys. Res., 103, 17099, doi:10.1029/98JD00900, 1998.

- 2694/01: How does one arrive at the "magic number" 1.5km?

  The choice of 1.5 km as the approximate ALH for carbonaceous aerosol that are not elevated was based on a sensitivity analysis showing that the in the UV the sensitivity to absorbing aerosol layers is below 2.5 km.

  However, as stated above, the OMAERUV retrieval reports an ensemble of height-dependent AOD solutions. The best-guess solution is the one associated with the CALIOP-based ALH (please see Page 2694 Line 5-10 in the manuscript). Thus the 1.5 km value is inconsequential to our study and is provided to the reader for completeness.

- 2694/03: Please explain where the different assumptions on vertical distribution come from for carbonaceous and sulphate aerosols.

  Change made.

  Page 10, Line 4: "The approximations for the shapes of the aerosol

extinction vertical profiles are based on ground-based lidar observations (Torres et al., 1998)."

- 2694/24: If CALIOP is still measuring today, replace *from* by *since*. Otherwise state the end of the CALIOP time period.

  - Change made, Page 10, Line 24.

- 2695/05: *ratio of aerosol 180-backscatter to extinction* is not clearly understand- able.

  Change made.

  Page 11, Line 1: "the ratio of aerosol backscattering to aerosol extinction"

- 2695/07: *level 2543nm aerosol extinction*?

  This is a type setting error. Page 11, Line 3 should read "…we used daytime CALIOP level 2 532 nm….".

- 2695/15: Please comment on the effect of assumed SSA on the extinction profiles.

  This is an internal retrieval assumption and an extended discussion is outside of the scope of this manuscript. However, a comparison of CALIOP 532 nm observations to ground-based LIDAR showed that the top and base height of aerosol and cloud layers of the two measurements generally agreed to within 0.1 km (Kim et al. 2008 – please see Page 2695 Line 25).

  Kim, S.-W., Berthier, S., Raut, J.-C., Chazette, P., Dulac, F., and Yoon, S.-C.: Validation of aerosol and cloud layer structures from the space-borne lidar CALIOP using a ground-based lidar in Seoul, Korea, Atmos. Chem. Phys., 8, 3705–3720, doi:10.5194/acp-8-3705-2008, 2008.

- 2696/08: Please state exactly which MODIS/Aqua data set you are referring to.

  Change made.

  Page 11, Line 30: "(MYD 14)"

- 2696/19: Here you write that Fig. 1 shows a 2006-2008 average; in the caption of Fig. 1, you talk about only the fire season of 2006-2008. Please be consistent.

  Change made.
  Page 12, Line 10: "averaged over the 2006-2008 fire seasons (July-November)"

- 2696/24: Please define what you mean by *spatial correlation coefficient*. If you simply took the Pearson correlation of the gridded data sets, then the use of "spa- tial" is

not justified, as this correlation coefficient does not really contain spatial information.

Change Made.

Page 12 Line 15: "The Pearson correlation coefficient of the two gridded 3-year averages"

- 2698/01: In which context did you *replace* OMAERUV with CALIOP ALH?

  Change made.

  Page 13, Line 18: "To do this we interpolated the OMAERUV AOD and SSA given on the 5 altitude nodal points to the CALIOP ALH".

- 2699/27: Further reasons for differences include RTM differences, possibly different aerosol parameters in the RTMs, . . .

  No aerosols are considered in the two radiative transfer calculations being compared here. The same doubling-adding method was used in the radiative transfer calculations.

  Please see Page 2699 Line 27 where we say: "The differences between the DOMINO and DISAMAR tropospheric AMFs in Fig. 6 represent … numerical differences that arise from higher resolution vertical layering in the DISAMAR radiative transfer calculations."

- 2702/08: Speaking of *cloud pressure [. . .] above [. . .]* is misleading; you could clarify by speaking of *higher clouds*.

  Change made.

  Page 17 Line 4: "the O_2-O_2 effective cloud is typically higher than"

- 2717: Please explain the meaning of *average [. . .] MODIS-Aqua active fires*.

  Change made.

  Figure 1 caption: "The average active fire number represents the 2006-2008 average number of observed daily active fires in each grid cell during the fire season."

- 2717: Doing the collocation based on a 0.5 degree radius means that effectively, your collocation radius is getting smaller towards the South . . .

  For the entire domain considered, the collocation radius would decrease from approximately 55 km at the top of the domain to approximately 48 km at the bottom. However, as the bulk of the observations occur in a

relatively narrow band in the middle of the domain, the effective difference in collocation radius is approximately 4-5 km. This will be refined in future analyses.

- 2718: It is not clear if the mean and std stated within the plot itself refer to only the CALIOP ALH (if so, what about OMAERUV ALH?) or the whole data set?

  The mean and standard deviation written in the plot are color coded in blue to correspond with the blue bars, which are labeled CALIOP ALH. We have added the following to the figure 2 caption to clarify.

  "The mean and standard deviation of the CALIOP effective ALH is 1.5 km and 0.62 km, respectively."

  Reporting a mean and standard deviation for the OMAERUV reported ALH would be misleading, as the ALH values for sulfate and non-elevated carbonaceous aerosol are discrete numbers. In other words, the probability distribution of the OMAERUV ALH is a combination of a binomial and continuous distribution. The appropriate estimate for the central tendency of the population would be the ALH value with the highest probability, which is apparent from the figure.

- 2721: Actually, the *solid* grey line is the least-squares fit.

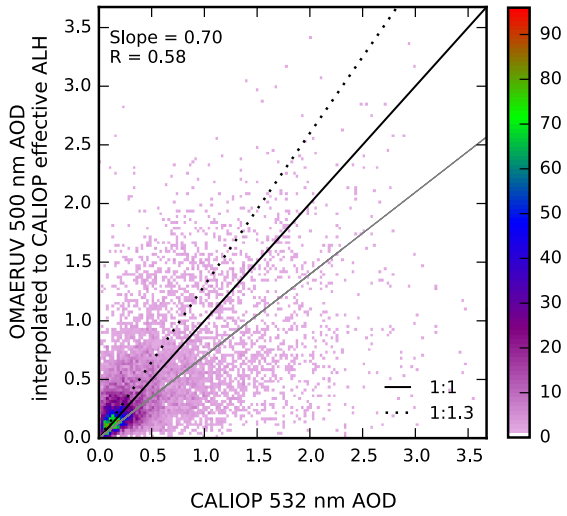
  Change made.

- 2721/2722/2723: Why do you use an additive "error bar" in Fig. 5 and a multiplicative "error bar" in Figs. 6+7?

  The gray dashed lines in Figs. 5-7 are not error bars. They are included in the figure as visual aids to the reader. However, the dashed lines delineating +/- 20% in Fig 6 & 7 are significant, as 20% is the reported estimate for the AMF uncertainty in the DOMINO retrieval.

  We have altered Figure 5 to also use multiplicative lines.

  New Figure 5:



- 2723: *AMFs* instead of *AMFS*

  Change made.

- 2724/2725: What are the horizontal black lines in the left plots (probably CTP, but that's not written in the legend as in the right plots)?

  Please see the captions for Figs. 8 & 9 where we state: "In all the plots, the O_2–O_2 retrieved effective cloud top pressure is shown as a horizontal black line, and the CALIOP effective aerosol layer pressure is shown as a dashed horizontal gray line."

- 2726: What are the dashed horizontal black lines in all plots?

  Change made.
  Figure 10 caption: "The dashed horizontal black lines are the 1.1, 1.0, and 0.9 horizontal grid lines."

- 2728: Please explicitly clarify that the solid line is the AL@850hPa, and the dashed lines are clouds.

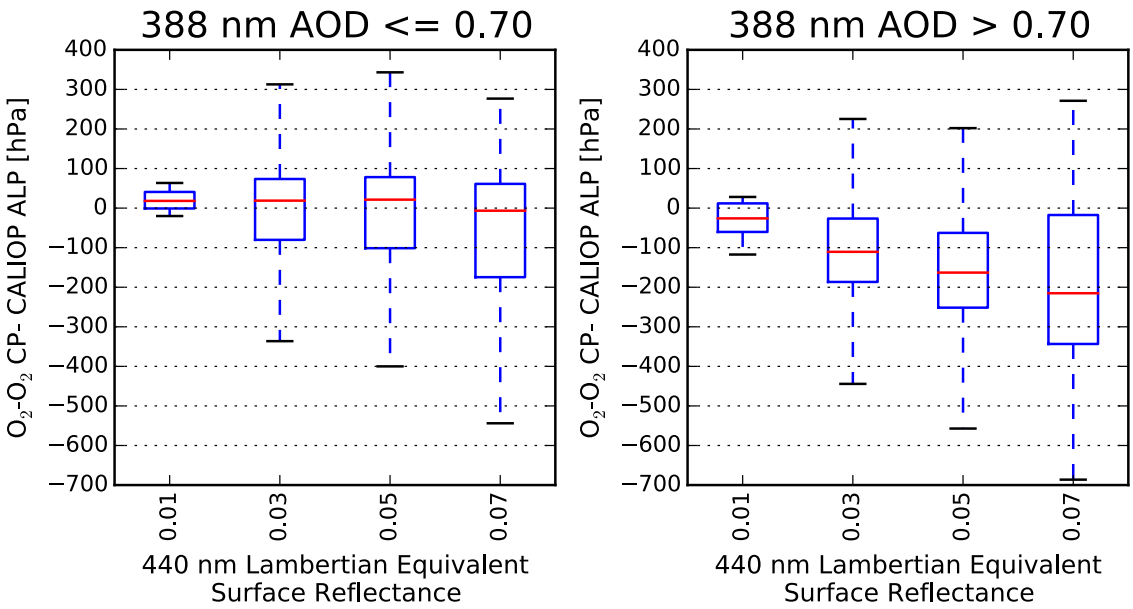
  Change made.

  Figure 12 caption: "Simulations of differential optical thickness for an aerosol layer centered at 850 hPa and extending for 300 hPa (solid red line). In each figure the differential optical thicknesses of Lambertian clouds with continuum reflectance equal to that of the aerosol layer simulation (i.e. equal cloud fraction) are shown for different cloud pressures (dashed lines)."

- 2729: Please speak of *surface spectral reflectance*, as albedo is a quantity aver- aged over all wavelengths. Also, please state the wavelengths of surface spectral reflectance and AOD.

  Change made.

  New Figure 13:



- 2731: In the discussion of the asymmetry parameter g (see p. 2709), it would be nice to discuss which parameter might be more realistic of biomass burning scenarios.

  Change made.

  Page 23 Line 23: “AERONET observations during the dry season in South America show that the average and standard deviation of the asymmetry parameter at 440 nm is 0.68 ± 0.02, with a range of 0.6 to 0.75 (Rosáraio et al., 2011; Sena et al., 2013).”