Response to reviewer #2

Thank you for the reviewer's effort to review our manuscript. During the revision processes of our manuscript, we re-wrote most parts of the manuscript. In addition, we added results of sensitivity tests and error analysis for additional aerosol parameters. During the revision, we changed the radiative transfer model to improve the interface of previous model for surface albedo. For this reason, we also revised the methodology to explain the new radiative transfer model and its condition

This paper explores the possibility to retrieve the aerosol altitude from the O2-O2 spectral band. This is a very important topic, in particular for trace gases retrievals from the UV and Visible spectral bands where there is a need to correct for aerosol effects. I agree with the comments written by Referee #1. The study presented here appears incomplete and gives rise to various questions about all the error sources which impact the quality of the retrievals. As highlighted by Referee #1, the presented study case does not allow to validate the proposed algorithm. Some sections include some confusing elements which need to be clarified (see below for details). Moreover, it is very hard to have a critical judgement and understanding of some results as some technical details are missing on the employed approaches for the analysis and the AEH algorithm.

→ In the revised manuscript, we added further details of our AEH retrieval algorithm and methodology for sensitivity test in Section 2 and 4. For error budget analysis, we added the aerosol particle size as an error source parameter in the Section 3.2.3.

Furthermore, we also retrieved other aerosol loading cases over East Asia for validation in Section 4 with the details of cases listed in Table 8. The algorithm was validated by using CALIOP data as a reference as shown in Figs. 13-15. Details of methodology for case study is described in Figure 12 and explained in the beginning of Section 4 as below:

"Figure 12 describes an AEH retrieval algorithm for the case study. In retrieving AEH, AOD is obtained from MODIS standard product (e.g., Levy et al., 2007). Although OMI aerosol product provides AOD at 500 nm, AOD from OMI was partially affected by aerosol height and suffered from cloud contamination due to its large footprint (Torres et al., 2002). For this reason, AOD from MODIS allocated to the OMI pixels as a reference AOD for the AEH retrieval. For type selection, the AE from MODIS and AI from OMI is respectively used for the information of size and absorptivity, to classify aerosol type into four following the method from Kim et al. (2007) and Lee et al. (2007). After determining AOD and aerosol type, LUT, which is generated as functions of geometries (SZA, VZA, and RAA), aerosol types and AODs, is used to determine the AEH information by using comparison between simulated and measured O4I value. The variables and their dimensions for the LUT calculations are shown in Table 7. Due to the limitation of the accuracy of aerosol type classification and those of AOD over land, this study estimates the AEH only over ocean surface. Although temporal and spatial variation of surface albedo influences the AEH result from error study, surface albedo is assumed to be a fixed value of 0.10, which is used in

sensitivity study. For case study, the LUT of O4I is developed by the aerosol model based on AERONET data over East Asia. Extensive AERONET dataset over East Asia are used to provide represent aerosol optical properties for the LUT calculation."

Finally, as this manuscript focuses on the feasibility to implement an algorithm, I wonder if a submission to Atmos. Meas. Tech. would not be more appropriate than ACP. Therefore, I suggest major revisions and clarifications for this paper before being submitted again. The most important revisions (addressed in detail in the following section) include: A complete and detailed description of the proposed algorithm and the employed approaches of analysis, in particular for the error analysis (Section 3.2) and for the DOAS analysis (Section 2.2); Clarification of the results and issues risen below: in particular about the error analysis, clarify please all the reference scenarios considered, how this can change depending on the variability of the geophysical conditions, and explain in detail the reason of the somewhat surprising small impact of surface albedo; Inclusion of more than 1 study case, or at least a more convincing case.

→ We improved the input interface of radiative transfer model. For this reason, RTM was changed from LIDORT to a newly developed radiative transfer model, VLIDORT. Details of VLIDORT and new methodology are described in Section 2.1. Details of methodology for DOAS are also additionally described on Section 2.2 in the revised manuscript with clear-sky comparison test.

After updating the model, we found that previous result for the impact of surface albedo was underestimated, thus revised all error budget and sensitivity studies for aerosol parameters and surface albedo. Furthermore, we additionally included the result about geometrical dependence as shown in Figure 8. Details are explained in Section 3.1.

Specific comments
Table 6 shows a summary of error sources and the total error budget for the AEH retrieval. The methodology of deriving this table should be described in more detail in section 3.2, using equations for example and giving a clear methodology. Moreover, the reference surface albedo should be given. We can expect that these numbers will change with respect to the geophysical conditions. Are these numbers based on a standard error propagation (i.e. assuming that each parameter will impact the result as random error)? If yes, the presented results may be somewhat underestimated.

→ We revised details of methodology in Section 3.2 with equation as below:
"Errors are also estimated in terms of key variables in the estimation of the O4I at 477 nm, with the variables and their dimensions as summarized in Table 3. For the error analysis of AEH retrieval, characteristics for all of extinction material are essential to consider. In this study, errors are analyzed in terms of AOD, aerosol vertical distribution, particle size and SSA for aerosol amount and properties. Surface albedo variation is also considered to represent surface condition. To estimate the error amount, the AEH error is converted from the half of O4I difference between adding and deducting perturbation of variables as shown in equation (1).

$$\varepsilon(Z) = \left| \frac{O4I(x+\delta x,Z) - O4I(x-\delta x,Z)}{2.0 \times dO4I/dZ(x,Z)} \right| \quad (1)$$

where $\varepsilon(Z)$ is the AEH error amount due to variable of error source, x, in AEH of Z, and δx is perturbation of AEH retrieval error source. The $\varepsilon(Z)$ value also depends on viewing geometries. Therefore $\varepsilon(Z)$ is represented for specific geometries together with averaging over all geometries."

→ The error study is basically estimated from radiative transfer model result. Therefore, the error analysis reflects result by changing target parameter only, while other variables are not changed.

Indeed, uncertainties on the AOD for example will likely result in a systematic error (i.e. bias) on the aerosol altitude. The evaluated uncertainty on the AEH retrieval induced by an error on the surface albedo of 0.02 appears surprising and should be explained (around 50 m for WASO case, less than 100 m for the other cases). It is much lower than the uncertainty due to the AOD (less than 200m) and SSA (between 229 and 2155 m). As explained in this paper, the AEH is strongly constrained by the O2-O2 SCD. However, this variable is also strongly driven by the O2-O2 continuum reflectance [Acarreta et al., 2004; Chimot et al., 2015] which, by definition, results from a combination of AOD (and associated additional scattering caused by aerosols) and surface albedo. Therefore, the surface albedo should be a key component (at least for a given range of AOT), and it is not understood why here this has so little impact. [Veihelmann et al., 2007] has also shown the importance of the knowledge of surface albedo for aerosol retrievals from the OMI spectral measurements.

→ After changing radiative transfer model, the AEH error due to surface albedo was redone, and the error budget results were also changed. Revised result is shown in Section 3.2.4 for surface albedo, and all error budget results are listed in Table 6 in the revised manuscript. From this result, the surface albedo is one of key factor for AEH estimation.

Finally, what is also missing is a theoretical discussion about the impact of clouds (e.g. in case of low cloud fractions). In the case of O2-O2 cloud retrievals, [Acarreta et al., 2004; Chimot et al., 2015] have shown that the effective cloud pressure value is very sensitive to the range of effective cloud fraction. For low cloud fraction, and thus low continuum reflectance and so low AOD, the relative variability of O2-O2 SCD is quite small and so it is more challenging to retrieve the aerosol altitude with a low uncertainty. The sensitivity of the AEH accuracy to AOD, and in general everything which impacts directly the O2-O2 continuum reflectance magnitude, should be discussed. It is expected that the AEH retrieval algorithm will be more accurate for large AOTs, while for low AOTs the AEH uncertainties should be higher.

→ This study focused on the aerosol retrieval over cloud-free pixels. For this reason, corresponding pixel is selected with extremely low cloud fraction value (0.02 for the case study in Section 4). In the revised manuscript, we added the result of AEH uncertainties as a function of reference AOD and AEH for respective error sources. Details are described in

Section 3.2 with Figs. 7 to 11 in the revised manuscript.

Table 8 presents the input parameters of the LUT used for the AEH retrieval. Following my comment above, it is not clear why the surface albedo is not one of the parameters. Does it mean this LUT is generated for one single surface albedo case (in line with the value given from OMLER over your study case)? Section 3 says that "the climatological value from OMI Level 3 (OMLER) is used in this study". How is it used exactly since no input parameter is present in Table 8? Same for surface pressure or surface altitude. What was assumed for these parameters? How does it impact the result of your study case? Section 2.2 (DOAS analysis) mentions that a factor of 1.25 is used as a correction factor on the O4 absorption cross section as suggested by [Irie et al., 2011; Lee et al., 2011]. However, such a factor is commonly employed for ground-based in- struments like MAX-DOAS (as done by these 2 papers). There is no explicit evidence this is needed in the general case of satellite measurements. Please explain why you considered it here. On the other hand, it is mentioned that such a factor should cover the temperature dependence of the O4 SCD. I do not think that such a scale factor can cover this effect in satellite measurements. The work by [Maasakkers et al., 2013] demonstrates that this dependence varies in time and space. This can have major impact on the effective cloud retrieval, in case of the O2-O2 cloud algorithm, mostly for cases with low effective cloud fraction (change in cloud pressure between 100 and 200 hPa). Impacts on the aerosol altitude retrieval should be investigated as well, and for different AOD. More literature review, where the impacts of aerosols on the effective cloud retrievals should be added e.g. [Castellanos et al., 2015; Chimot et al., 2015; Lin et al., 2013; Lin et al., 2015], as they analysed the impacts on the O2-O2 spectral measurements.

→ To reflect your comment, we did not use the correction factor value after revising manuscript. In section 2.2, we described the details of surface albedo assumption. Instead, the surface albedo is assumed to be 0.10, because frequent Lambertian equivalent reflectance (LER) is larger than minimum LER as described in Kleipool et al. (2008). In the revised manuscript, we revised in Section 2.2 as below:

"Figure 3 shows the comparison of the O_4 SCD at 477 nm from a look-up table (LUT) with the dimension as in Table 2 against OMCLDO2 for aerosol and cloud free pixels in year 2005. The clear sky region is selected for the Pacific Ocean with cloud fraction less than 0.02 from OMI observation. The surface albedo is assumed to be 0.05, which is similar to the minimum Lambertian equivalent reflectance (LER) over clear ocean surface (e.g., Kleipool et al., 2008). Because the standard product of the O_4 SCD is only estimated at the 477 nm band, the results can be compared only at this band. To minimize the DOAS fitting error, the observed data from OMI is selected by the fitting precision less than 2% and the quality flags for spectral fitting and pixel condition are also considered. As shown in Figure 3(a), the correlation coefficient of determination (R^2) is 0.864 with a slope of 1.050, and the LUT exhibits a ratio of 0.86±0.05 to the values obtained from OMI standard values. Despite the statistically significant R^2 and slope values between the two values, there exists negative bias by about 14%.

The bias between the retrieved from LUT and estimated from standard product values can be attributed to the differences in the O_4 cross section data and the lack of their temperature and pressure dependence as noted from the previous works by Wagner *et al.* (2009), Clemer *et al.* (2010), and Irie et al. (2015). For this reason, ground-based measurements adopted the correction factors to cross section database. However the bias effect for the cross section difference is limited as shown in Figure 2, and the correction factor for the cross section database in the previous studies cannot be adopted to the space-borne measurements. From Kleipool et al. (2008), the minimum LER is defined to be the 1% cumulative probability threshold, and frequent LER value is typically higher than minimum LER over clear ocean, although cloud screening was perfectly executed before LER calculation. To account for the difference between simulated and observed SCD, the LUT was re-calculated by changing condition to the surface albedo of 0.10. The corrected result is shown in Figure 3(b), where the R^2 is 0.865 similar to that before the correction, but the negative bias is removed to 0.98±0.05 and the regression line slope is 1.123. Although the comparison result is not perfect, the calculation by the VLIDORT simulates the satellite observation and can be used for sensitivity tests to retrieve aerosol height."

→ Although correction factor of O4 is still challenging issue for satellite and ground observation, we used the method to modify surface albedo.

We also added the literature review in Section 1 in the revised manuscript as below:
"For OMI measurement, the O_4 band at 477 nm has been widely applied to estimate cloud information (e.g., Accarreta *et al.*, 2004; Sneep *et al.*, 2008). Especially, the cloud information retrieved by O_4 band at 477 nm was used for air mass factor (AMF) analysis with the consideration of aerosol optical effects for the NO_2 column retrieval (e.g., Castellanos et al., 2015, Chimot et al., 2015; Lin et al., 2014; Lin et al., 2015). Although O_4 absorption band around 477 nm varies also due to cloud existence, it can be also used for the aerosol optical parameter estimation. Veihelmann *et al.* (2007) introduced that the 477 nm channel, which locates major O_4 band, significantly adds degree of freedom for aerosol retrieval by using principal component analysis, and Dirksen *et al.* (2009) adopts the pressure information obtained from OMI O_4 band to identify a plume height for aerosol transport cases. "

As Referee #1 pointed out, appropriateness of the selected study case is questionable. It is mentioned that the AEH derived from OMI is performed for a scene at 1.5 deg (i.e. around 150 km) away from the LIDAR site. This is a very long distance. The comparison with CALIOP seems to present some large differences (1.-1.5 km for CALIOP vs. 2.6 +- 1.7 km for AEH). More or different study cases should be presented for a more robust comparison.
Furthermore, Section 2 mentions that OMI data are selected with cloud fraction fraction less than 0.02. [Chimot et al., 2015] and [Boersma et al., 2011] have shown that the OMI effective cloud fraction is very sensitive to aerosols and AOD, and can reach values between 0.1 and 0.15 for AOT = 1. However values of 0.02 may indicate very little aerosols present in the OMI data (AOT likely less than 0.2).
→ For the validation study, we added the comparison results in multiple aerosol loading

cases. Furthermore, we directly compared to the AEH from CALIOP, and presented the result in Figs. 13-15. Cases for Figure 15 are listed in Table 8 in the revised manuscript. To estimate the AEH from OMI, we used the MODIS AOD after collocating OMI pixel, because of its low sensitivity to aerosol vertical distribution. Detail of method is described in Figure 12. If co-located MODIS AOD is not shown in specific OMI pixel, we did not estimate the AEH, although cloud fraction from OMI is lower than 0.02. By restricting those criteria, most of cloud effect can be neglected because spatial resolution of MODIS is better than those of OMI.

Technical corrections
P. 7934, 2-4: "using simulated radiances by a radiative transfer model, ... (LIDORT), and ... (DOAS) technique". Please separate LIDORT and DOAS techniques in this statement. Here, DOAS could be understood as a model name, not as a retrieval technique.
→ We separate the methodology of VLIDORT and DOAS in the revised manuscript.

P. 7934, 13-14: "knowledge on the aerosol vertical distribution type": please reformulate. Do you mean aerosol vertical distribution and aerosol type?
→ We revised the paragraph "assuming knowledge on the aerosol vertical distribution shape".

P. 7934, 25: "in regional and global scale": replace "in" by "at"
→ We revised on the paragraph "at regional and global scale" in the revised manuscript.

P. 7935: The necessity to know aerosol layer height for trace gases retrievals should be mentioned too.
→ We revised on the sentence "The information on the aerosol layer height is important, because the variation of the aerosol vertical distribution affects radiative process in the atmosphere near the surface and trace gas retrieval for air mass factor calculation." in the revised manuscript.

P. 7935, 12: Change "Vertical structures" to "Vertical profiles"
→ We changed in the revised manuscript.

P. 7935, 25: "CALIOP haS been successful (not "have")
→ We changed in the revised manuscript.

P. 7937, 12-14: Please specify that this refers to the impact of aerosols on the O4 signal. Reformulate "path length of light" as "length of the average light path".
→ We changed in the revised manuscript.

P. 7940, Section 2.2.: Please give more details about the approach implemented for the DOAS retrievals, based on the WinDOAS software. In particular, specify what you mean by "using a non-linear least squares method". Some equations with the retrieval state vectors

and considered / assumed elements would help the reader.

→ We referred the reference of WinDOAS software for methodology. In addition, we revised Section 2.2 to explain the details of methodology.

P7940, 22-23: "comparison of the 477 nm O4 SCD between the inversion from a LUT": which LUT are you refereeing here? No LUT is explained before in the manuscript. And there is no use of a LUT usually to derive the O4 SCD.

→ We mentioned the sentence for the clear-sky LUT calculation in the revised manuscript as below:

"Figure 3 shows the comparison of the O_4 SCD at 477 nm from a look-up table (LUT) with the dimension as in Table 2 against OMCLDO2 for aerosol and cloud free pixels in year 2005."

Recommended additional literature

Acarreta, J. R., De Haan, J. F., and Stammes, P.: Cloud pressure retrieval using the O2-O2 absorption band at 477 nm, J. Geophys. Res., 109, D05204, doi:10.1029/2003JD003915, 2004. 8388, 8399, 8400, 8402

Castellanos, P., Boersma, K. F., Torres, O., and de Haan, J. F.: OMI tropospheric NO2 air mass factors over South America: e_ects of biomass burning aerosols, Atmos. Meas. Tech. Dis25 cuss., 8, 2683–2733, doi:10.5194/amtd-8-2683-2015, 2015. 8389, 8408.

Chimot, J., Vlemmix, T., Veefkind, J. P., de Haan, J. F., and Levelt, P. F.: Impact of aerosols on the OMI tropospheric NO2 retrievals over industrialized regions: how accurate is the aerosol correction of cloud-free scenes via a simple cloud model?, Atmos. Meas. Tech. Discuss., 8, 8385-8437, doi:10.5194/amtd-8-8385-2015, 2015.

Lin, J.-T., Martin, R. V., Boersma, K. F., Sneep, M., Stammes, P., Spurr, R., Wang, P., Van Roozendael, M., Clemer, K., and Irie, H.: Retrieving tropospheric nitrogen dioxide from the Ozone Monitoring Instrument: e_ects of aerosols, surface reflectance anisotropy, and vertical profile of nitrogen dioxide, Atmos. Chem. Phys., 14, 1441–1461, doi:10.5194/acp-25 14-1441-2014, 2014.8388, 8389, 8408.

Lin, J.-T., Liu, M.-Y., Xin, J.-Y., Boersma, K. F., Spurr, R., Martin, R., and Zhang, Q.: Influence of aerosols and surface reflectance on satellite NO2 retrieval: seasonal and spatial characteristics and implications for NOx emission constraints, Atmos. Chem. Phys. Discuss., 15, 12653–12714, doi:10.5194/acpd-15-12653-2015, 2015. 8409.

→ We added the recommended literature in the revised manuscript.