This document contains the responses to the editor's comment on the manuscript version 2, followed by a version 3 manuscript (starts in page 8). The reviewers' comments and questions are in bold. For each comment / question, the authors' reply / answer is in black, and the corresponding modifications in the manuscript version 3 are marked in blue color.

Author's response to the editor

Thank you very much for your comments on our manuscript. In the followings please find responses to your comments.

Your manuscript # amt-2018-40 has received comments and suggestions from two anonymous reviews for which you have just submitted a response addressing their concerns. Thank you. Along the review process, I have read your paper and gave some thoughts on the research content. As I understood, the concept of the retrieving aerosol SSA in the near-UV region by constraining AOD quite resembles to the methods presented in Satheesh et al. (2008) and Gassó and Torres (2016). Both earlier studies, and the results of your own paper demonstrate that it is possible to retrieve aerosol SSA and layer height from the color ratio information (UVAI) in the near-UV given the AOD as an a priori and a pre-defined aerosol model. It was a surprise that your paper completely misses to discuss and cite these two papers relevant to the present study.

We have read the two research papers we missed out upon that you mention in the comment. We appreciate the studies you suggested here (the publication of Satheesh et al. was actually published in 2009). From our view, the goals and / or the methods of these two studies do not resemble to ours. Satheesh et al. (2009) provided a hybrid retrieval method by combing OMI and MODIS measurements, and Gassó and Torres (2016) aimed to discuss the discrepancy between OMAERUV AOD and other independent measurements. But both studies did discuss the relation among aerosol layer height, aerosol concentration, and aerosol absorption, thus we have included them in the introduction part.

Satheesh et al. (2009) used a hybrid approach to retrieve aerosol layer height (ALH) and aerosol single scattering albedo (SSA). They combined the OMI aerosol product (OMAERUV), which is sensitive to ALH and aerosol absorption, with MODIS's accurate aerosol optical depth (AOD), which is insensitive to ALH. Their study has a similar object as ours, that is to retrieve SSA from satellite measurements, but the method is not the same. Both studies retrieve ALH and SSA from given a priori aerosol models. Satheesh et al. used MODIS AOD as the parameter to constrain the operational OMAERUV retrieval, while we use the absorbing aerosol index (AAI). The role of AAI used in Satheesh et al. (2009) is a qualitive parameter to distinguish absorbing aerosols from non-absorbing ones.

Specifically, Satheesh et al. (2008) extrapolated MODIS AOD to the near UV band. Using this MODIS-produced AOD to constrain the standard OMI AOD inversion procedure (OMAERUV) allows to derive improved ALH and SSA (that specified in the LUT). In our study, we used MODIS standard AOD at 550 nm to constrain the radiative transfer calculation in the forward simulation, and used OMI measured AAI to constrain the backward retrieval of ALH and SSA. Furthermore, Satheesh et al. (2009) only compared the SSA retrieved with the hybrid-algorithm with that retrieved with the standard OMI algorithm SSA. They did not validate it with measurements from other instruments such as AERONET.

Satheesh et al. mentioned the difficulty of extrapolating MODIS AOD from visible band to 388 nm where OMI requires the AOD information for retrieval (especially difficult for small particles, e.g. biomass burning aerosols). They first applied a linear least square fitting (log-log scale) for AOD as a function of wavelength, then they improved the method by including information on the AOD spectral curvature. The relation between UV and visible AOD may provide some clues to determine the relation between UV and visible SSA and refractive index, and it is worth to study in a separated research. Satheesh et al. also mentioned that measurements of aerosol absorption in the UV spectral range are rare, which makes it difficult to validate the retrieval results. We met similar difficulties in determining the spectral dependence of aerosol properties from

the visible to the UV band. But most importantly, we did not notice that the use of MODIS AOD can eliminate the uncertainty of ALH assumption. We have included this as one of the reason that use MODIS AOD to constrain the forward simulation.

Gassó and Torres (2016) compared the OMAERUV aerosol product with independent measurements (MODIS and AERONET) in order to: (1) assess the quality of the OMAERUV AOD retrieval over ocean (section 3); (2) estimate the impact of cloud contamination on their AOD retrieval (section 4); (3) demonstrate the effect of variations in aerosol concentration and height on OMAERUV by comparing the operational OMAERUV algorithm and the OMI-MODIS joint algorithm presented in Satheesh et al. (2009) (section 5); (4) determine whether the assumed aerosol models that leads to discrepancies in AOD retrieval of section 5 (section 6). Gassó and Torres (2016) aimed to evaluate the influences of several factors on the AOD retrieval, rather than to retrieve the SSA from measured AAI (with MODIS AOD constrain). Through case studies, Gassó and Torres (2016) evaluated the hybrid retrieval method presented by Satheesh et al. (2009) via case studies. At high AAI magnitude (AAI > 1.8), the retrieved ALH is comparable with CALIOP measurements, although their method is quite sensitive to small variations in the input AOD in case of low AAI. Although they seemed to discuss the interplay among AAI, ALH and AOD, the AAI was not used as the constraint to retrieved aerosol absorption. However, Gassó and Torres (2016) concluded that AAI alone cannot be used quantitatively if no ALH or AOD information is available. This is consistent with our finding that the accuracy of the retrieved SSA depends strongly on the uncertainties in AOD and ALH.

In summary, although these two studies discussed the interplay among AOD, ALH, and aerosol absorption (SSA / AAI / refractive index), but the purpose / method differ from ours. We use the measured AAI as an additional constraint to retrieve SSA, which is not the case the other two studies. But both studies provided useful information in aerosol research that we can include in our manuscript. Here are the corresponding contents that we have included study of Satheesh et al. (2009) and Gassó and Torres (2016):

In section 1 introduction: But this aerosol absorption over near-UV is highly sensitive to the assumption on aerosol layer height. Satheesh et al. (2009) therefore used the τ from MODerate-resolution Imaging Spectroradiometer (MODIS), which is independent of aerosol layer height, to constrain the OMAERUV retrieval. The validation showed that compared with operational OMAERUV algorithm, the retrieved aerosol height by the hybrid method is in a better agreement with air-borne measurements, implying a potential improvement in aerosol absorption retrieval. This OMI-MODIS joint retrieval was also evaluated by Gassó and Torres (2016). They found under less absorbing condition, the hybrid method is sensitive to the variations in the input τ, which is used to select the retrieved pair of aerosol layer and ω_0. (line 43 - 49)

In section 3.1.2 MODIS and OMI aerosol optical thickness: Besides, the MODIS retrieved τ is free from the uncertainty triggered by assumed aerosol profile (Satheesh et al., 2009). (line 187 - 188)

In section 3.2 Methodology: Previous research suggested AAI cannot be quantitatively used without τ or z_{aer} information (Gassó and Torres, 2016). (line 257 - 258)

In this inversion process, your study employs an aerosol model developed using the AERONET data. It was not clear in the Methodology section what kind of spectral dependence is assumed between the two near-UV wavelengths. A number of studies, using laboratory measurements and satellite data, have shown that the carbonaceous aerosols generated from the open field agricultural burning and wildfires exhibit a strong wavelength-dependent of absorption (imaginary part of the refractive index) in the near-UV region. In our earlier study (Jethva and Torres, 2011, ACT), we learned that the relative spectral dependence in the imaginary part of the refractive index needs to be increased to 20% with reference to no spectral dependence (equivalent to black carbon) in order to obtain a better

agreement in AOD and SSA between OMI and AERONET. A strong wavelength dependence in aerosol absorption in the UV has been shown as a proxy for the amount of organics in the biomass burning smoke.

The color ratio (UVAI) information in the near-UV strongly varies with the Absorption Angstrom Exponent, a parameter that quantifies the spectral dependence of aerosol absorption. Since your paper lacks to provide this essential information, it is hard to interpret the SSA retrievals presented in the paper. Perhaps, a significant disagreement of ~0.06 in SSA between the OMI retrievals and that of AERONET reflects the issue described above that you should consider while revising your manuscript. Please verify the relative spectral dependence in absorption assumed in the aerosol model and discuss/mention in the revision.

Apology for the unclear explanation in the Section 3 Methodology and datasets. Indeed, the spectral dependence of aerosol absorption has a significant influence on the retrieval of SSA. As you mentioned in Jethva and Torres (2011), compared with the gray aerosol assumption used in OMAERUV algorithm, an aerosol model with wavelength dependence can significantly reduce the bias in retrieved AOD and achieve better agreement of retrieved SSA with AERONET measured SSA (70% retrieved data is within +/-0.03 difference).

Therefore, one has to be careful when relating the aerosol refractive index in the UV to the one in the visible band. Jethva and Torres (2011) finally adopted a spectral dependence with a 20% increase in the 354 nm imaginary refractive index with respect to the value at 388 nm (absorbing Angstrom Exponent around 2.5 to 3.0 for seven carbonaceous aerosol models in OMAERUV). It is an improvement compared to the operational application, where the aerosol properties are pre-assumed. However, aerosol properties evolve over time and thus also over space. Also, even for a specific aerosol type, the aerosol properties vary from one study to another. Thus, we prefer to directly use AERONET measurements to define the aerosol type rather than to use to one of the pre-defined aerosol types.

The corresponding content in the manuscript has been rephrased into: The spectral bands of the AERONET instrument at this site only cover the visible band. Due to the absence of observations, assumptions have to be made on the spectral dependence of aerosol properties to obtain their values in the near-UV band. The properties of biomass burning aerosol depend on the type of fuel, the procedure producing the smoke, the age of the smoke, and also the atmospheric conditions (Reid et al., 2005). Using measurements to constrain the input aerosol refractive index can reduce the uncertainties due to a-priori knowledge. Our treatment on the complex refractive index is as following: (1) take the complex refractive index at visible band (440 to 675 nm) from AERONET measurements; (2) linearly extrapolate the complex refractive index to near-UV band. The real part n_r for radiative transfer calculation is obtained in this step. A slight wavelength dependence of n_r is found from the measurements; (3) for the imaginary part n_i, we multiply it (for the entire wavelength from UV to visible) with a scaling factor as we treat it as a free parameter. By varying the value of the scaling factor, both the magnitude and the wavelength dependence of n_i can change to meet the requirement of retrieval (line 234 - 243).

We have now also included the retrieved refractive index in Table 2. In our case the spectral dependence between 354 and 388 nm is less than 5%, which is smaller than the 20% in Jethva and Torres (2011). We also added a plot to show the original AERONET measured (dashed line) and retrieved complex refractive index (scaled by a factor) in Fig.9. Although the scaling factors can amplify the spectral dependence, it is not very large due to the flat character of the original refractive index. We have added an explanation for this: There are many sources accounting for this discrepancy. First of all, the nearest site Santiago_Beauchef is not exactly in the primary biomass burning regions as mentioned in section 3.1.3. The AERONET site is located in downtown, where reflective urban or industrial aerosols may have been mixed with the smoke enhancing the ω_0. This would also affect the complex refractive index used in radiative transfer calculation, since we use the AERONET measured refractive index to constrain the forward simulations. According to Table 2, the retrieved n_i reveals that the difference between 354 and 388 nm is less than 5%. This small spectral dependence of n_i is mainly determined by AERONET measurements in the visible band (dashed lines), whereas the effect of the scaling factor is minor in this case (Fig.9). We thus find a much weaker wavelength dependence than in the Jethva and Torres (2011) study, where a 20%

difference between the two UV wavelengths was applied to OMAERUV algorithm to achieve the result that 70% of the retrieved ω_0 differ less than ± 0.03 from the ω_0 from the AERONET measurements. A stronger spectral dependence of n_i between 354 and 388 nm would allow simulations to reach a higher AAI while keeping n_i at a relatively low level, so would retrieve a higher ω_0 at 550 nm. The presence of non-absorbing aerosols weakens the spectral dependence (particularly in the UV spectral range) and the linear extension would overestimate the aerosol absorption. In this situation, the uncertainties due to assumed spectral aerosol properties might compensate the measurement errors to some extent (line 313 - 326).

In our case, we use the AERONET measurement to constrain the spectral dependence of aerosol properties, but it might be improved if observations from other observational aerosol properties in UV band would be available as constraint. This is summarized in the conclusion: This study proves the potential of utilizing OMI measured AAI to quantitatively characterize aerosol optical properties like ω_0. Currently, it is challenging to retrieve and validate results without reliable aerosol profile information. In the future, the availability of daily global aerosol profile data, e.g. the L2 aerosol layer height product TROPOspheric Monitoring Instrument on-board Sentinel-5 Precursor (TROPOMI) that is underdevelopment (Sanders and de Haan, 2016), are expected to provide a stronger constraint on the forward calculation and to significantly reduce the uncertainty in the retrieved aerosol properties. Perhaps, more sophisticated assumptions on spectral-dependent aerosol absorption (e.g. steeper gradient of n_i in UV than visible band) have to be made and evaluated by other observational aerosol properties in UV spectral range, e.g. AERONET measured τ in UV band, instead of only depending on measured refractive index in visible band (lie 381 - 388).

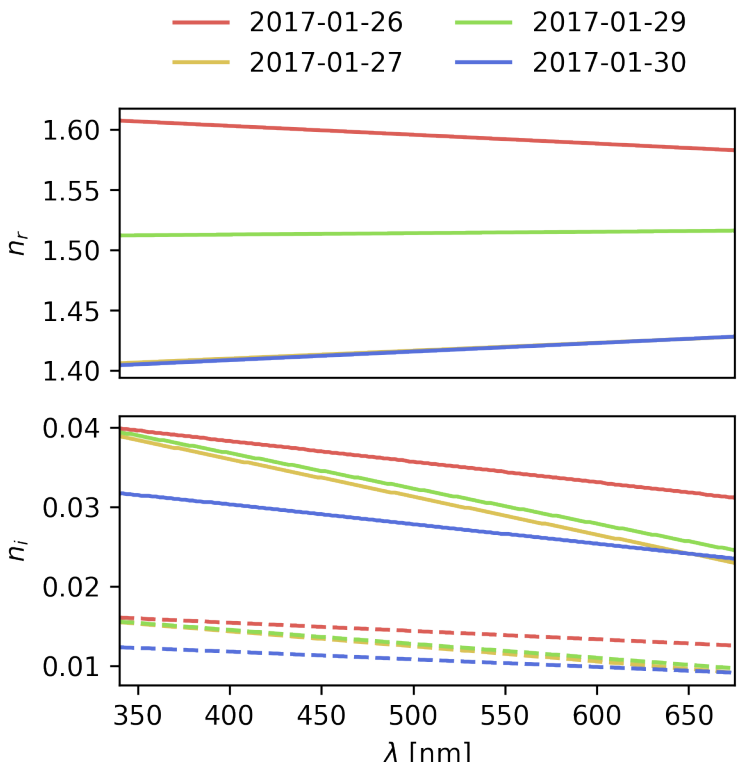

Figure.9 Retrieved complex refractive index for each case. The dashed line in lower panel is the wavelength dependent imaginary refractive index (n_i) measured by AERONET.

Table.2 Summary of retrieved results (applying IQR outlier detection).

	Date	2017-01-26	2017-01-27	2017-01-29	2017-01-30
AAI	AAI median (OMAERO)	2.52	2.38	4.05	2.61
	AAI median (DISAMAR)	2.17	2.48	3.81	2.49
	Relative difference (%)	-13.88	4.20	-5.93	-4.60
	RMSE	0.67	0.51	0.60	0.41
Aerosol profile	z_{aer} [km]	4.9	4.5	4.7	4.7

	Δz [km]	2			
n_i	n_i at 354 nm	0.0395	0.0382	0.0388	0.0314
	n_i at 388 nm	0.0386	0.0366	0.0373	0.0306
	N_i difference between 354 and 388 nm	2.33%	4.37%	4.02%	2.61%
ω_0 at 550 nm	ω_0 (AERONET)	0.89	0.89	0.92	0.91
	ω_0 (DISAMAR)	0.83	0.81	0.87	0.85
	Relative difference (%)	-6.74	-8.99	-5.43	-6.59