RC – Referee comments are in Black
AR – Authors response are in Blue

Reviewer – 1

This is a paper that addresses a new approach to the remote sensing (RS) of aerosol absorption by satellite, and with comparison to the same ground-based RS retrievals that are a major input to the algorithm/technique. The use of all AERONET retrieved parameters except for imaginary refractive index is justified for the retrieval of SSA in the UV wavelengths, which is currently not an AERONET product (440 nm is the shortest wavelength in V3). The concept of the paper is somewhat new in scope and has potential to add significantly to the current knowledge on global aerosol absorption. However, there are numerous significant shortcomings to this paper, especially related to the estimated uncertainty of these satellite retrievals of aerosol single scattering albedo (SSA). The uncertainty of the satellite retrievals is somewhat inconsistent in the text and the values in Table 2. You need to clearly state that the uncertainty in SSA from these retrievals due to the complete ensemble of the known sources of error. These known sources of error are uncertainties in all of these factors: AOD, BRDF, aerosol layer height (ALH), size distribution, real refractive index and satellite sensor calibration uncertainty. Full uncertainty calculations and a complete discussion seems to be missing from this paper, and it is essential. Your assumed uncertainty in surface albedo of 0.01 is quite small and is somewhat hard to believe, except in the UV where the reflectivity is quite low for most surface types. References are missing to support this level of surface reflectance uncertainty for the visible wavelengths. Additionally surface albedo is not what is important here but it is BRDF that is a function of both view and solar zenith angles. Additionally, since you use a climatology of size distribution and refractive index from AERONET how much does that affect the retrievals at high and/or low AOD when there are departures from the presumed averages used in your climatological values? See comments below in 'Specific Comments' about the dynamic nature of size distributions as a function of AOD and relative humidity. Also, you make no mention of the magnitude of the uncertainty in satellite sensor calibration and yet this needs to be included in your calculation of overall uncertainty of the satellite retrievals. Your computed uncertainty of +-0.03 in the UV is as good as AERONET at 440 nm (see Sinyuk et al. 2020) and is somewhat hard to believe since for AERONET the effect of uncertainty in surface reflectance and ALH is minimal at AOD(440)>0.4, while the factor of ALH is quite a large of error in the OMI satellite algorithm retrievals in the UV. Table 2 does seem to include AOD uncertainty calculations as +- 0.02 but you fail to discuss this in the text. Also the header of Table 2 only mentions the effects of ALH, so this header needs to be re-written to include the other two factors.

Another problematic aspect of this paper is classification of some sites as biomass burning sites (ex. Arica, Chile) when that this clearly erroneous and other sites as urban aerosol types when they are clearly biomass burning sites (ex. Cuiaba, Brazil). I give details below (in Specific Comments) including several other sites plus some references in the peer-reviewed literature regarding the aerosol types at these AERONET sites.

Additionally, the referencing of published scientific literature is sometimes lacking and sometimes completely erroneous in this manuscript. I provide numerous examples below in my Specific Comments. The authors need to follow normal standards for correct referencing for a journal of high standards such as AMT, and in this manuscript these standards have not been met.

As a result, my opinion is that this manuscript requires extensive revisions and corrections before it may possibly be suitable for publication in AMT.

Specific Comments:

RC: Line 40: It would be nice to quantify here the SSA value corresponding to 50% less UV flux. Also, it seems that you meant 'decrease' instead of 'increase' in this sentence.
AR: Corrected as 'decrease'.

RC: Lines 51-53: It should also be mentioned that most in situ measurement techniques make several assumptions and have significant 'corrections' made to the data to overcome instrumental shortcomings or issues. Therefore the community in general does not consider these data to be a 'gold standard' for validation and there currently is no such gold standard data product available.
AR: We concur with reviewer's comment. The insitu measurements, either taken in lab or measured in the fields, often undergo corrections and subsequent releases. We have added a sentence mentioning instrumental assumptions and corrections.

RC: Lines 60-63: This statement is not entirely accurate. The Hybrid scan has been taken for several years now from newer model Cimel instruments utilized by AERONET. These Hybrid scan retrievals are robust down to 25 degrees solar zenith angle as compared to 50 degrees for the almucantar scans (see Sinyuk et al. 2020). Therefore the high SZA limitation no longer applies. Also, these same instruments have taken 380 nm sky radiance data which is now being analyzed by the AERONET group for a future SSA retrieval at this wavelength. Although this data product is not yet available it should be mentioned that the data exits at many sites for a 380 nm retrieval of SSA, which is an ongoing research topic within the AERONET group. Additionally the recently published paper of Sinyuk et al. (2020) provides uncertainty values of SSA for all AOD levels (figures and tables) so that a data user can assess the level of uncertainty of these retrievals at the 4 retrieval wavelengths for a complete range of AOD for four different aerosol types.
AR: Revised description of the AERONET products. We have now clearly mentioned about the availability of 380 nm sky radiance data and the expected 380 nm SSA in the future upgrades of the AERONET inversion product.

RC: Lines 76-77: SSA is sometimes assumed constant for a region in some MODIS retrievals, but not one constant value globally. You should reference the relevant papers on this regionally varying assumed SSA by Remer et al. 2005 and Levy et al. 2007.
AR: Added a sentence mentioning the use of regionally varying constant SSA in MODIS-DT aerosol products.

RC: Line 90: 'Sahara belt' is an odd terminology. Maybe you mean the Saharan and/or Sahelian regions?
AR: Corrected as 'Saharan' region.

RC: Line 102: This is confusing, as MODIS does not measure at a local noon overpass time. Do you mean 'local time' here instead of 'local noon'?
AR: Corrected as 'local time'.

RC: Line 120: You need to reference Giles et al. (2019) here for the AERONET Version 3 Level 2 AOD data that are cloud screened and QA checked.
AR: Included appropriate reference for AERONET Version 3 Level 2 AOD product.

RC: Line 121: You say nine nominal wavelengths and then you only give 8 wavelengths, omitting 1640 nm.
AR: Corrected for 9 wavelengths.

RC: Line 122: You say the AERONET measurement interval is 15 minutes, but for many instruments the current interval is 5 minutes and has been for a few years. Please convey this in the text.
AR: Corrected for the AERONET measurement time interval.

RC: Line 122: You need to say how you interpolated the spectral AOD measured by AERONET to the OMI measurement wavelengths. A second order polynomial fit of AOD versus wavelength in logarithmic coordinates is much more accurate than using the Angstrom or linear fit (Eck et al., 1999).
AR: Added description on how the AERONET τ is converted to satellite wavelengths in the revised manuscript (section 3.3.1).

We derive the τ at the satellite wavelengths using the closest available measurement (within ±2 hours centered at the satellite overpass time) and α through the power-law approximation (Ångström, 1929) as shown in the below equation. For OMI wavelengths, the AERONET 340 and 380 nm measurements are readily available while τ_{354} is obtained with λ_{Ref} = 380 nm and $\alpha_{\lambda\_Ref} = \alpha_{340\text{-}440}$. For the few sites with older models of AERONET photometer that does not have direct sun measurements at 340, and 380 nm (for example Banizoumbou, Avignon, etc.), we use λ_{Ref} = 440 nm and $\alpha_{\lambda\_Ref} = \alpha_{440\text{-}675}$. Similarly, for MODIS wavelengths the τ at 466 and 646 nm are obtained using λ_{Ref} = 440 nm and $\alpha_{\lambda\_Ref} = \alpha_{440\text{-}675}$.

$$\tau_\lambda = \tau_{\lambda_{Ref}} \left(\frac{\lambda}{\lambda_{Ref}}\right)^{-\alpha_{\lambda\_\lambda Ref}}$$

RC: Line 123: Dubovik and King (2000) is the retrieval algorithm paper and not did analyze the AOD measurement accuracy. Therefore, this is a totally inappropriate reference here. The AERONET paper that provides an analysis and estimate of AOD measurement uncertainty is Eck et al. (1999).
AR: Eck et al. (1999) citation for AERONET AOD uncertainty is now included.

RC: Line 128: The Dubovik et al. 1998 paper is a totally inappropriate reference here. Please replace this with Dubovik et al. (2006).
AR: Dubovik et al. (2006) reference added.

RC: Line 129-130: This is a completely obsolete reference for the uncertainty of SSA from AERONET. For Version 3 retrievals there are spectral estimates of SSA uncertainty in Sinyuk et al. (2020) for each of the 4 wavelengths that are also provided as a function of AOD.
AR: Corrected for appropriate reference.

RC: Lines 130-132: This is confusing since below and in Table 1 you say that you use L2 retrievals, yet in this sentence the mention of L1.5 data implies that possibly some of this data may also have been used in this paper. Please clarify this issue in the text.
AR: Corrected for clarity. AERONET Level 1.5 data products are not used in this work.

RC: Lines 135-137: Note that you have an error in Table 1 where you say that for AERONET retrievals Version 2 is used, while in the text you say Version 3 is analyzed.
AR: This is not an error. We have now mentioned it clearly in the text as well. AERONET version 3 level 2 almucantar inversion products were released in late 2017, much latter than we started working on this project. Therefore, for constructing climatology of aerosol particle sizes AERONET version 2 level 2 data is used. Other than that, elsewhere (i.e., AOD inputs for the retrievals and SSA comparison) in our work version 3 level 2 products are used.

RC: Lines 164-167: In section 2.3 MODIS, it seems that at least one reference for MODIS data is warranted in the text.
AR: Appropriate reference is now included.

RC: Line 177: Did you create a size distribution climatology that is a function of AOD? It is well known that fine mode size increases significantly as AOD increases at many sites due to aerosol aging and/or hygroscopic growth. This has been shown in many AERONET papers, see Eck et al. 2012 where the fine radius increases by over 50% over the range of AOD at the GSFC site. Additionally, Eck et al. 2010 presents examples where the fine mode radius doubles over a wide range of fine mode fraction.
AR: We used the entire range of AOD to create seasonal climatology of size distribution. The revised manuscript includes sensitivity tests on our SSA retrieval by perturbing the particle sizes (volume mean radius, VMR) for all aerosol types by 20%. We used 20% perturbation based on examination of the seasonal climatology of particle sizes as a function of AOD and most frequently occurring AOD bin size for over several sites.

RC: Line 211: This sentence is very confusing, please clarify in the text here. Do you average all AERONET measurements of AOD within the +-2 hour interval? Also average all retrievals of size distribution and refractive index within the +-2 hour interval? What specifically do you mean by 'keep it intact'?
AR: Rewritten the sentence for clarity.

- We look for valid AERONET AOD measurements within ±2 hours of satellite overpass and assign the AOD closest in time to all ground pixels. No average of AOD measurements is performed in this work.
- To develop LUT of radiances, we use the entire record of AERONET particle sizes available in the inversion product without any temporal constraint.

RC: Line 221-222: How accurate is this aerosol typing? How many types are there to choose from? Please give a short summary of the typing procedure and it's accuracy or reliability.
AR: In the revised manuscript, aerosol type information is described in section 3.3.2.
"We use a combination of Extinction Ångström Exponent (α_{440_675}) derived from AERONET and near UV Aerosol Index (UVAI) from OMAERUV product to categorize the observed aerosols into three basic types – dust, carbonaceous, and urban/industrial. Initially, our algorithm

uses $\alpha_{440\text{-}675}$ to identify the aerosols as coarse ($\alpha_{440\text{-}675} \leq 0.2$) and fine ($\alpha_{440\text{-}675} \geq 1.2$) mode dominated particles. Threshold $\alpha_{440\text{-}675}$ of 0.2 chosen for coarse mode particles unambiguously identifies dust aerosols. However, the sample of fine mode particles consists of both absorbing carbonaceous and weakly absorbing urban type aerosols. The near-UV aerosol index is an excellent indicator to identify the presence of absorbing aerosols. Threshold UVAI value adopted from OMAERUV algorithm is used to separate carbonaceous (UVAI ≥ 0.8) and urban (UVAI < 0.8) aerosols, respectively."

RC: Lines 224-226: Please provide a short description of the accuracy of this aerosol height climatology in the text, as this is a critical factor in the overall uncertainty of the satellite retrievals of SSA in the UV wavelengths.
AR: A brief description of monthly aerosol layer height climatology derived from the joint OMI-CALIOP data set is provided in the section 3.3.2. The expected uncertainty in the ALH from the prescribed OMI-CALIOP data set is within ±1 km (Torres et al., 2013).

RC: Line 237: What is the wavelength of the AOD in the x-axis of Figure 4? This should be added to the figure.
AR: Revised the figure to add τ_{440} in the x-axis.

RC: Lines 239-241: It should be noted that these are significantly different SSA values than those retrieved by AERONET in the visible wavelengths for the GSFC site. See Giles et al. (2012) that gives SSA at 440 nm = 0.96 and SSA at 675 nm = 0.95. These are VERY large differences (~0.06 to 0.10), especially when it is considered that the SSA parameter range is only ~0.80 to 0.99 for >99% of AERONET retrievals. However you show a large trend as a function of AOD and the lowest AOD bin may be dominating these averages. Why give equal weight to low AOD retrievals that you are aware have very large uncertainty and likely large biases? It is good to see that the Table 3 values for SSA are for AOD(440)>0.4 and that these are much closer to the AERONET retrieval values of the Level 2 database.
AR: The average SSA values reported here is for the entire range of τ. However, we have now replaced it with average SSA for observations with $\tau_{440} > 0.4$ and mentioned it in the main text.

RC: Lines 252-253: This sentence is somewhat confusing and suggests that you may possibly have used different surface reflectances from the MAIAC product that you documented above.
AR: We used MAIAC MCD19A1 spectral BRF or surface reflectance product. Brief description of the product along with the reported uncertainty estimates are provided in the section 3.3.3.
"The prescribed measurement-based uncertainty in the MCD19A1 product ranges from 0.002 – 0.003 for visible wavelengths (Lyapustin et al., 2018)".

RC: Line 260: This site name has been mis-spelled, it is Mauna Loa, the Langley calibration site for AERONET. Please note that the AOD is also VERY low at these 3 sites, well below the threshold for accurate retrievals of SSA. Additionally, the Mauna Loa site is on a high mountain (3400 meter elevation for the site) so the AOD from AERONET would not correspond to the aerosol signal of the total atmospheric column as measured by OMI or MODIS. You should have filtered out all sites on mountains for this reason before attempting this analysis.
AR: Revised manuscript now excludes the AERONET sites with very low AODs.

RC: Line 270: Angstrom must always be capitalized as it is the name of a scientist.
AR: Corrected as 'Ångström'.

RC: Line 275: Why not reference the Angstrom 1929 paper, as it is the origin of this parameter.
AR: Included the reference for the Ångström exponent.

RC: Line 282-284: The selection of AE<0.2 for coarse aerosol cases is somewhat extreme as there are relatively few cases of desert dust with AE<0.2 in the AERONET database. Utilizing AE<0.4, which is still coarse mode dominated, would have resulted in many more dust cases to analyze. There are many papers based on in situ measurements that have identified fine mode dust thus the AE of airborne desert dust does not often equal zero or have a negative value which would be the case if there were only coarse mode particles. The 0.2<AE<1.2 bin for mixed mode cases encompasses a very wide range of fine mode fraction of AOD, ~30% to 70% at 500 nm (see Eck et al. 2010).
AR: We agree, the choice of AE ≤ 0.2 for dust aerosols might be extreme. However, allowing the AE up to 0.6 as dust aerosols will only (a) increase in the number of dust samples, and (b) likely introduce 'mixtures' of aerosols in that category – especially in the Sahel region. Therefore, we chose to use the AE ≤ 0.2 to derive unbiased spectral signature of coarse mode dominated dust aerosols.

RC: Lines 287-294: The factors you have identified here (a-d: (a) aerosol extinction measurements, (b) estimation of particle size distribution, (c) real part of refractive index, (d) calibration of satellite measured TOA radiances) however are significant sources of uncertainty in the satellite retrieval of SSA. How much do these factors affect the uncertainty of your SSA retrievals with this algorithm?
AR: The revised manuscript now includes sensitivity tests for the estimated error in SSA retrieval due to all input variables used in the algorithm.

RC: Lines 309-311: How did you arrive at an uncertainty estimate of 0.01 for surface reflectance? This is a very small uncertainty in my opinion since surface reflectance varies seasonally (vegetation phenology), and also as a function of view and solar zenith angle within a day. Additionally, how did you arrive at +-1 km for uncertainty in Aerosol Layer Height?
AR: For the UV wavelengths, we use a surface albedo data set developed from long-term measurements using the minimum Lambertian Equivalent Reflectance (LER) – directly adopted from the operational OMAERUV product. The prescribed uncertainty for this surface albedo data set is expected to be within ±0.01 (Torres et al., 2018). For surface characterization in visible wavelengths, we use MAIAC MCD19A1 BRF product. The prescribed measurement-based uncertainty in the MCD19A1 product ranges from 0.002–0.003 for visible wavelengths (Lyapustin et al., 2018). However, in our sensitivity tests we used a consistent ±0.01 perturbation in surface reflectance for all wavelengths.

To obtain an estimate of ALH required for the retrieval of SSA for both carbonaceous and dust aerosols, we use joint OMI-CALIOP data set. The joint OMI-CALIOP data set uses coincident observations and aerosol index to identify absorbing aerosols and obtain corresponding CALIOP derived layer height from backscattering profiles at 1064 nm. The prescribed uncertainty in the

derived layer height primarily stemming from limited sampling of CALIOP overpasses is expected to be within ±1 km (Torres et al., 2013).

RC: Lines 312-313: Why is the SSA satellite retrieval uncertainty expected to increase with increasing wavelength, just because AOD is less at larger wavelengths or some other reason? If it is related to AOD only then the dust cases would not show a decrease in SSA uncertainty for the longer wavelengths. Please clarify in the text.
AR: Revised the error analysis to include uncertainties due to all input variables in the algorithm. "Among the nine input variables used in our algorithm, the $\Delta\omega_o$ at 340 nm arises mostly from (in descending order) the uncertainties in calibration of TOA radiances, sub-pixel cloud contamination, ALH, particles sizes and so on. While for the visible wavelength at 646 nm, the $\Delta\omega_o$ arises mostly from (in descending order) cloud contamination, surface reflectance, calibration of TOA radiances, particle sizes and so on. These sensitivity tests clearly indicate that $\Delta\omega_o$ is (a) spectrally dependent due to multiple variables, (b) decreases with increasing τ, and (c) varies with absorbing nature of aerosols".

RC: Lines 316-318: "Our analysis shows that for small τ440 (~0.2), the error in retrieved SSA is much higher (> +-0.05) for visible wavelengths, while that in the near-UV region reaches up to +-0.03." Again, is this because the AOD is higher in the UV wavelengths? I am surprised that the ALH effect, which is much greater in the UV, does not counter the AOD wavelength dependence. If it is AOD wavelength dependence that is the main factor then this statement is not true for cases with low values of the Angstrom Exponent.
AR: Please see the above response.

RC: Lines 326-328: The way you have written this section it appears to me that you have only considered the factors of uncertainties in ALH and surface reflectance combined in your estimates of SSA uncertainty. It seems that you have not considered the effects of 0.01 uncertainty in AOD as measured by AERONET and this does not even factor in the spatial variance of AOD over the OMI pixel size. Satellite sensor calibration is also not mentioned in your written description of these estimates of SSA uncertainty. What value of satellite sensor calibration uncertainty did you use? Did you include all of these sources of uncertainty in your calculations but just failed to document them in the text of this paper, or vice versa?
AR: Revised the error analysis to include uncertainties due to all input variables in the algorithm.

RC: Line 344: The aerosols at the Arica site are definitely not biomass burning aerosols as you suggest here. They are dominated by sulfate emissions from copper smelters and therefore non-absorbing. See Eck et al (2012) for a discussion of fine mode size and SSA for this site as follows: "...typical of most retrievals at Arica, where the average SSA is 0.98 for all wavelengths, from nearly 400 retrievals from 1998 to 2000, where AOD (440 nm) >0.4. This is consistent with the principal aerosol sources in the Arica region, as the SO2 emissions from copper smelting create sulfate particles that are non-absorbing"
AR: The AERONET site 'Arica' mentioned in the section 5.1.1 is a typo/mistake. For the regional average SSA presented in figures 6, 7, 8, and 9 the sites names are provided in the upper right corner with abbreviations D (dust), M (mixed), C (carbonaceous), and U (urban) indicating the aerosols type that were averaged to produce the regional spectral SSA. In the revised manuscript these figures are re-produced, and the sites names are mentioned more clearly.

RC: Line 345: Note that Sao Paulo is a major urbanized region (one of the largest on Earth) and therefore the primary aerosol type is urban-industrial, not biomass burning.
AR: Yes, we agree. Please see the above response, in the figure 6 Sau Paulo is denoted with 'U' indicating urban aerosols.

RC: Lines 357-359: Why do you even include a discussion of urban aerosols in this section since this section is titled 5.1 Biomass Burning? Have you averaged the Arica retrieval results into the urban or into the biomass-burning category in this confusing paragraph? Note that the Arica site is neither in an urban region nor is it a biomass-burning site. In my opinion the Arica, Sao Paulo and CEILAP-BA sites should be dropped from this subsection, or you should rename the title of section 5.1.
AR: The section 5 is re-written to avoid confusion.

RC: Lines 360-362: Please note that the higher absorption at the CUIABA sites are due to biomass burning of Cerrado vegetation (similar to wooded savanna) which exhibits more flaming phase combustion therefore more BC than the predominately smoldering combustion at the tropical rain forest sites (see Schafer et al 2008). Your lumping the CUIABA site aerosol type into the urban category of Sao Paulo is erroneous.
AR: In our results we observed both smoke and urban aerosols for the Cuiaba site (please see figure 6 where Cuiaba is denoted with 'C' and 'U'). As mentioned, carbonaceous and urban aerosol types are distinguished using a threshold near-UV AI – synonymous to the aerosol categories in OMAERUV algorithm. Based on extensive tests on the OMI signal strength on all surface types it is determined that a minimum UVAI of 0.8 is required to identify absorbing aerosols (Torres et al., 2007, 2013). Although UVAI is an excellent indicator to identify absorbing aerosols, the large OMI pixel size and sub-pixel contamination of signal strength might at times underestimate the UVAI placing the observed aerosols in urban category. In the revised discussion of carbonaceous aerosols over South America, we have included appropriate references for the high absorption of smoke at Cuiaba.

RC: There are many errors in Table 3 for South America in my opinion. You list the Rio Branco site as urban when it is located in a rural region dominated by biomass burning emissions. The Arica site is missing from Table 3 so why even include it in the averaging in your analysis in this section? You also list a high percentage of retrievals for all of the biomass burning sites in South America as urban which is erroneous. Why do you classify some smoke cases as carbonaceous and some as urban in Table 3? This apparent mis-classification needs to be discussed in the text.
AR: We revised the section 5 (sub-titles) and analysis to include only sites with well know aerosol sources from the literature. However, we chose to keep the results reported in Table 3 (Table S1 in revised version) intact. Please refer to the above response.

It should be noted that our technique to retrieve SSA for carbonaceous and urban aerosols differ only in the use of ALH and uses the same site-specific LUT radiances. It is well known and shown in our sensitivity tests that the effect of ALH is pronounced in UV and gradually reduces towards the visible spectrum. Nonetheless, the results reported in Table 3 provide useful information on aerosol absorption over these sites.

RC: Line 365: Again, it is an odd choice to mix this Pretoria site (urban) in with two rural sites that are dominated by biomass burning aerosols, in a section titled Biomass Burning.
AR: Revised the section 5 (sub-titles) and analysis to include only sites with well know aerosol sources from the literature.

RC: Lines 365-366: There are very few natural forest fires in this southern Africa region. Most biomass burning emissions are from savanna burning initiated by farmers or livestock grazers with minor contribution of crop residue burning. Please refer to Eck et al. (2001 & 2003) for discussion of biomass fuel types in southern Africa.
AR: Added appropriate references for biomass fuel types in southern Africa.

RC: Line 374-375: It seems very doubtful that these relatively high AOD cases in southern Africa are urban aerosol dominated although there may be some mixtures. This urban versus carbonaceous type classification is very confusing and needs to be addressed before these regions are analyzed/introduced in these sections.
AR: Please see above responses regarding the aerosol types of categories based on EAE and UVAI used in this work.

RC: Lines 377-378: Please give the spectral range of AAE here for these values and also note that for the visible to NIR spectral region the AAE of urban or biomass burning types is never as high as 2.2 in the published literature.
AR: In the revised manuscript only carbonaceous aerosols (section 6.1 – revised version) over Southern Africa are reported for Mongu, and Skukuza. Average $AAE_{340\text{-}646}$ for these carbonaceous aerosols are 1.7 and 1.6 during JJA and SON. However, as per the aerosol type identification scheme employed here there are few aerosol observations categorized as urban aerosols (Table S1).

For carbonaceous and urban aerosols where black carbon is the sole absorber the AAE values are close to 1 at UV to NIR spectral range. However, aerosols with organic compounds the AAE values varies widely up to 6 (Bergstrom et al., 2007; Kirchstetter et al., 2004 and the references therein) in the UV spectral range, and greater than 1 in the UV-Vis spectra. In addition, it should be noted that AAE values in the literature are derived from linear-fit of power-law expression for any spectral range, while the AAE determined in this work are computed based on the AAOD values at the particular wavelength pairs – these two approaches also lead to some differences.
The mean AAE values reported in this work for carbonaceous (Figure 14) and urban aerosols (Figure 16-in revised version) are consistent with those in the literature.

RC: Lines 384-385: In northern Australia it is not just savanna but also woodland and forests.
AR: Added this information.

RC: Lines 396-397: Please be clear here, are these values AAE. The text should be easier to read without constantly referring to the Figure.
AR: Yes, it is UV-Vis spectral dependence of aerosol absorption ($AAE_{340\text{-}646}$).

RC: Line 397: Note that neither of these 2 sites (Jabiru and Lake_Argyle) are strongly influenced by urban aerosols therefore this classification seems erroneous.

AR: The revised text is now limited to carbonaceous aerosols observed over the Jabiru and Lake Argyle sites.

RC: Line 402: Note that the Saada site is adjacent to the city of Marrakesh (~1 million pop.) therefore influenced by urban emissions while the Tamanrasset site is in a rural region in the middle of the Sahara.
AR: Although the site Saada is located adjacent to the city, dominantly coarse-mode dust aerosols are observed during JJA and SON months.

RC: Lines 422-425: This is a completely erroneous reading/interpretation of Eck et al. (2003). There have never been smoke particles in Africa with AE ranging from 0.2 to 0.5 from AERONET measured AOD spectra. You need to eliminate this false statement. Smoke/dust mixtures may have a wide range of AE but these mixtures cannot be called 'smoke particles'. Also note that Eck et al. (2003) reports no AOD measurements or retrievals of smoke properties from West Africa. This paper is focused on Southern Africa biomass burning aerosols, a completely different region. The inaccurate and misleading use of references in this paper is somewhat disturbing. From Eck et al. (2001), from page 3442 of the ZIBBEE paper: "Liousse et al. [1995] found AE (computed for similar wavelengths: 450, 650, and 850 nm AOD) for savanna burning smoke to range from 0.84 for aged smoke to 1.42 for fresh smoke at Lamto, Ivory Coast. However, in that West African location it is possible that these relatively low AE values may be influenced by the presence of Sahelian/Saharan coarse mode dust as a second aerosol type."
AR: We mentioned in the previous line that these aerosols ($0.2 < \alpha_{440\text{-}870} < 1.2$) are mixture of dust and carbonaceous aerosols. In the next line, it is a typo/mistake and should have been 'mixture' of aerosols. In the revised manuscript aerosols with mixed mode ($0.2 < \alpha_{440\text{-}870} < 1.2$) particles are eliminated.

RC: Lines 429-433: See Eck et al. (2010) for data and discussion of the highly absorbing smoke cases at the Ilorin site, from grassland and treed savanna biomass burning aerosols in the winter season (DJF). Include some comparison in the text.
AR: Added appropriate information with reference.

RC: Line 438: This section should be renamed to "Middle East/North Africa/Arabian Peninsula' since only 1 site out of the 4 sites you have listed is actually geographically located on the Arabian Peninsula (per Wikipedia).
AR: Renamed as 'Middle East'.

RC: Line 459: Classifying the aerosol at Missoula and Rimrock sites in the urban/industrial category (section 5.3 here) is somewhat absurd. These are far from having significant sources of urban/industrial pollution and just about all of the cases with moderate to high AOD (>0.4 at 440 nm) could readily be attributed to biomass burning sources.
AR: Please see above responses regarding the aerosol types of categories based on EAE and UVAI used in this work.

RC: Line 473: No, there is no similarity between the aerosol sources of the sites you analyze in your section 5.3.1 Western North America with the aerosol sources / types in Eastern North

America. As mentioned above 2 of the 3 sites you list for western NA are dominated by biomass burning aerosol sources.
AR: Removed this sentence.

RC: Line 477-478: You should mention that 4 of the 5 sites in this subsection are in a relatively small area, the central mid-Atlantic US (3 in central MD including GSFC), while one site is in the mid-West US (Bondville, Illinois).
AR: The revised analysis now includes regions: central US (Sioux Falls, BONDVILLE), Mideast US (SERC, GSFC, MD_Science_Center) for urban aerosols.

RC: Line 482-484: This statement is not clear, please elaborate here to better explain what you are referring to.
AR: Revised this sentence, to clearly mention that 'for weakly-absorbing aerosols the error in SSA retrievals is high due the identified factors from our analysis such as cloud contamination, surface reflectance, particle sizes and so on'.

RC: Line 487: There are 17 sites that are averaged together here for Europe, much more than for any other region. Some discussion regarding differences between sites is warranted. Also it seems probable that the somewhat noisy looking wavelength dependence of SSA for the SON season is likely due to a small sample size. The number of days of observations should be included in these plots, Figures 6-9, and when the sample size is small and therefore statistically weak it should be noted.
AR: Added some description of the aerosols over the European sites. Since these figures are seasonal averages of several sites over a region it is difficult to indicate the sample size of each site. The readers are advised to refer Table S1 for the sample size.

RC: Line 492-493: However this organic carbon absorption that you suggest is inconsistent with the AAE values in the same plot, as these AAE values which are close to 1 are typical values associated with black carbon absorption. Please discuss this apparent discrepancy and explain why you interpret such an AAE value as being associated with organic carbon absorption. Also include references to support your interpretation.
AR: We agree, AAE values close to 1 are associated with black carbon absorption. Revised sentences(s) consistent with the obtained results.

RC: Line 504: This section is very odd, in that you only include one site and this site is one of the largest urbanized regions in the world (Mexico City) that also happens to be located in Central America. Mid-Atlantic North America, which is your title of this subsection, is Maryland, Virginia etc. Mexico City is equally distant from the Pacific Ocean and Caribbean Sea therefore using mid-Atlantic to describe its location is beyond strange. It seems that the authors are either extremely careless with geographical labeling (or very careless in writing) or need to become more familiar with commonly used regional/geographical names.
AR: Replaced the subtitle Mid-Atlantic North America with Mexico City, Central America.

RC: Line 509-510: "It is clearly evident that such absorption curve and seasonal variation is a result of prevailing mixture of aerosols." This is a confusing sentence. Please elaborate what you

mean here by a mixture of aerosols since elsewhere you define an AE range as mixtures due to fine/coarse particle size mixtures.
AR: In the revised manuscript, the typical spectral SSA expected for the dust, smoke and urban aerosols are clearly described with references before any results are presented. The spectral SSA noted for urban aerosols over the Mexico City deviates from the typical behavior indicating mixture of aerosol components from high vehicular emissions and other sources.

RC: Line 512-514: Please discuss the uncertainty in your computed values of AAE somewhere in this paper. The AAE parameter is highly susceptible to small errors in AOD and SSA and the uncertainty in both AAE and AE increases as the wavelength range decreases due to the resulting small differences in AOD that approach the uncertainty level in the AOD itself. The uncertainty in the AAE for 354 to 388 nm is therefore very high for this reason of relatively small differences in AOD between these two close wavelengths (only 34 nm apart).
AR: Yes, AAE is highly susceptible to small errors in AOD and SSA. We added a section to discuss the uncertainties in the computed AAE by perturbing the SSA at 0.01 intervals.

RC: Line 518: This reference makes no sense as there is no mention of Mexico City in this particular paper (Eck et al. 1998). The use of inappropriate or plain wrong references in this paper is disconcerting at best. The authors need to follow normal standards for correct referencing for a journal of high standards such as AMT.
AR: Removed this reference.

RC: Line 521-522: Please note in the text that for cases of AOD(440)>0.4 as shown in this figure the two sites in China (Beijing and Xianghe, only ~60 km apart) completely dominate the statistics of these 4 site averages. The AOD in these China sites are very high while the AOD levels for the two sites in Japan are much lower such that there are relatively few cases in Japan that exceed the AOD threshold of 0.4. Averaging multiple sites in these types of plots can sometimes be justified but in this case it is very misleading.
AR: The revised analysis now includes regions: northeastern China (Beijing, XiangHe), Japan (Shirahama, Osaka).

RC: Line 522: If you want to label this Figure 9b plot 'Eastern China' then leave the 2 sites in Japan out of the data averaging. In fact, you write this entire section as though the Japanese sites are not included so why did you average these data in with the Chinese sites data at all?
AR: Please see the above response for northeastern China and Japan sites.

RC: Line 525-526: Note that the variation of AAE as a function of fine mode fraction (FMF) for both the Xianghe and Beijing sites are shown in Eck et al. (2010). For low FMF which is equivalent to your dust category, the AAE for both sites was ~2.5 which is consistent with most AAE values in for dust aerosol in the published literature. However your dust value of AAE is much lower than that and therefore you should explain why it is anomalously low for this aerosol type.
AR: From Eck et al., (2010) - Figure 17 c & d, the AAE for coarse particles noted over XiangHe and Beijing are 2.37 and 2.73, respectively. In this work, the AAE noted for dust (EAE $\leq$ 0.2) over northeastern China is 1.56 for 340-646 spectral range.

It is likely that these coarse particles are influenced by black carbon components over such highly polluted environments. However, due to large particle size (average $\alpha_{440\text{-}870}$ ~0.09) the spectral SSA noted still shows increasing SSA with wavelength. Among the regional dust observations presented in this work, northeastern China exhibit high absorption in visible wavelengths (ω_o ~0.93 and 0.95 at 466 and 646 nm). Chaudhary et al (2007) reported insitu measurements of coarse mode particles over XiangHe during March-2005 that exhibit high absorption in visible wavelengths (ω_o ~0.70-0.94 at 450, 550 and 700 nm). Li et al., (2007) explained the variation in SSA for coarse particles during March-2005 over XiangHe is a result of synoptic fluctuation – passage of cold fronts that uplifted ground-level pollution to higher altitudes influencing the aerosol absorption.

Similar low $AAE_{340\text{-}646}$ values for coarse mode ($\alpha_{440\text{-}870} \leq 0.2$, dust) are noted for few sites over the Sahelian region during DJF (burning season). It is likely that these coarse particles over Sahel are influenced by black carbon amounts emitted from biomass burning.

RC: Line 534-535: "The spectral behavior of urban aerosols is similar to carbonaceous aerosols with decrease in magnitude of average SSA, AOD, and UV-Vis AAE." This sentence seems to be somewhat incomplete, as what decrease in these 3 parameters are you referring to? A spectral decrease? Please clarify what you mean here.
AR: Removed this sentence.

RC: Lines 559-561: I have not been convinced that your separation of urban versus carbonaceous aerosol types is robust or has much relation to other classifications in the published literature. There is a lack of discussion of the accuracy of the separation of these two fine mode aerosol types.
AR: The aerosol classification scheme employed in this work uses combined Extinction Angstrom Exponent (EAE) derived from AERONET and near UV aerosol Index (UVAI) derived from OMAERUV product. The EAE and UVAI threshold values for the three aerosol types are: Dust – EAE $\leq$ 0.2, Carbonaceous – EAE $\geq$ 1.2 & UVAI $\geq$ 0.8, and Urban – EAE $\geq$ 1.2 & UVAI $<$ 0.8, respectively. It should be noted that although UVAI is an excellent indicator to identify the presence of absorbing aerosols, due to large OMI footprint and sub-pixel contamination of signal strength might at times underestimate UVAI and categorize the observed aerosols in urban category.

RC: Lines 578-581: However, you inexplicably have averaged the rainforest burning dominated sites with the cerrado vegetation burning dominated site (CUIABA). Note that cerrado vegetation is much like wooded savanna in southern Africa and that is why the SSA values for Cuiaba are lower than for the other South American biomass burning sites, see Schafer et al. 2008. You also have made a distinction between carbonaceous and urban types for these SA biomass burning sites which does not make any sense except for the Sao Paulo site (which has very few retrievals with AOD(440)>0.4). In short Figure 6 is quite misleading since you have mixed apples and oranges so to speak and then average the whole ensemble of sites.
AR: Yes, the regional average spectral SSA for carbonaceous aerosols over South America is derived from observations at Alta Floresta, Cuiaba, Ji Parana and Santa Cruz. Here our purpose is to derive regional average SSA that could be used for guidance in selection of absorption models for satellite aerosol retrievals.

In the revised manuscript, the figures showing regional average SSA are clearly displayed with the sites names to avoid confusion.

RC: Line 585: This would make more sense to add 'the savanna grasses' to 'in the central region' here.
AR: Added 'the savanna grasses'.

RC: Line 588-589: Biomass burning is always a mixture of flaming and smoldering combustion, but when the fraction of flaming combustion increases then the black carbon production increases and the SSA decreases. Please correct this sentence since you imply that this Australian vegetation burns entirely in the flaming phase, which is false.
AR: Corrected as 'biomass burning happens dominantly through flaming phase……'.

RC: Line 595: Again, the uncertainty in AAE for such a narrow wavelength range is much greater than for the 358 to 646 nm range. You should (as a computational exercise) vary the AOD by +0.02 at 354 nm and by -0.02 at 388 nm and see how different the AAE is for this expected AOD uncertainty alone. Add to this the uncertainty in SSA and it may not be very clear if the AAE is really that different in the UV alone from the UV-Visible wavelength range values, or just within the uncertainty error bars.
AR: Uncertainty in AAE for the three wavelength pairs is now included in the revised manuscript.

RC: Lines 597-598: "flaming combustion prevails" is too strong here as there is just a higher fraction of flaming combustion, while smoldering still produces more than half of the smoke aerosol.
AR: Replaced 'flaming combustion prevails' with 'where the contribution of flaming phase combustion is high'.

RC: Line 599: Please note that for the Ilorin site, Eck et al. (2010) found AAE to vary from 1.37 for fine mode dominated to 2.1 for dust dominated as a function of fine mode fraction. Please put your results in that context of variation as a function of Angstrom Exponent or FMF.
AR: We have now mentioned it clearly that the fine mode ($\alpha_{440\text{-}870} \geq 1.2$) carbonaceous aerosols found over Ilorin has average $AAE_{340\text{-}646}$ 1.38.

RC: Line 603-605: Mixing with coarse mode aerosol is only a part of the explanation of lower AE in these sites. Another factor that you should mention is that the fine mode particle size is larger at these sites due to aging processes of coagulation, condensation, hygroscopic growth and cloud processing.
AR: In the revised manuscript we limited the discussion of spectral SSA in the main text to only coarse-mode dust, fine-mode carbonaceous, and fine-mode urban aerosols. We eliminated the discussion of intermediate-mode particles ($0.2 < \alpha_{440\text{-}870} < 1.2$) in the main text and reported these results only in the Table S1 – supplemental material for informational purposes.

RC: Lines 610-612: I would suggest that you change 'often found' to 'sometimes found' since this varies greatly depending on the continent, region and season.
AR: Done.

RC: Lines 612-614: The reference of Torres at al. (2002) for this phenomenon (change in desert dust SSA during transport over the Atlantic from north Africa) is not very robust since the very large TOMS pixel size is very susceptible to partial cloud contamination which would be much more of an issue over the Atlantic Ocean than over the desert source regions and would thus yield higher SSA over the ocean. Additionally, the aerosol height utilized in these retrievals may have introduced significant uncertainty that may also differ for these two regions.
AR: Removed this sentence.

RC: Lines 615-618: Your suggested explanation seems very unlikely to be true. See in Eck et al. (2010) the section on the Ilorin site where the fine mode (biomass burning) aerosols are highly absorbing in the Sahel/Sudanian zones since this is primarily grassland and savanna burning with a relatively high contribution from flaming combustion (more BC produced). The SSA decreased at this site as more fine mode smoke was mixed with the dust. Additionally, some desert dust aerosol sources that advects into the Sahel region are relatively weakly absorbing. The Bodele Depression, which is perhaps the largest single dust source, is an example of weakly absorbing mineral dust since some of the material is diatomaceous sediment, which does not contain iron oxides. See Eck et al. (2010) and Di Biagio et al. (2019) for more information on the SSA of this specific source and other dust sources.
AR: Re-written the sentence for clarity and added appropriate references.

Lines 618-619: These dust SSA values from your retrievals should be compared to the values in the literature such as Di Biagio et al. (2019).
AR: Added appropriate references and comparison of dust SSA.

RC: Line 620: Please include in the Figure 11 caption an explanation of the 5.2 and 6.2 with the X inside the circle symbol.
AR: The circle and the X symbol was used to denote break in the bar chart when the AAE exceeds the maximum value in the y-axis (AAE). The revised version now replaces bar charts showing regional AAE with box-plots showing lower and upper quartile range of observations.

RC: Lines 625-628: It should be noted that this is such a wide range of AAE for dust (1.5 to 3.5) that it could be argued that even with very large retrieval uncertainties you still can fall within this range. Please defend the value in these dust AAE retrievals including estimates of the uncertainties.
AR: The range of mean $AAE_{340\text{-}646}$ values reported in the summary section correspond to all regions where dust aerosols ($\alpha_{440\text{-}870} \leq 0.2$) are observed in our sample. Excluding northeastern China and Sahel, the mean $AAE_{340\text{-}646}$ noted for remaining regions fall within the range 2.2-3.7 (Figure 12 and 15 – revised version). Considering the uncertainties in our AAE estimates these values agree well with those in the literature.

RC: Lines 630-632: Same comment as immediately above: It should be noted that this is such a wide range of AAE for dust in the literature that it could be argued that even with very large retrieval uncertainties you still can fall within this range.
AR: Revised manuscript includes discussion on the uncertainties in the computed AAE.

RC: Line 638: Please include in the Figure 12 caption an explanation of the 5.1 with the X inside the circle symbol.
AR: Revised manuscript now shows the same data as box plot with central 25 and 75-percentile range of observations.

RC: Line 665: Please provide the number of days of data for each site in Figure 13 (for both the AERONET and satellite retrievals) so that the relative statistical robustness may be evaluated.
AR: Revised figure now shows the number days and observations of the data.

RC: Line 666-667: Please note in the text that the uncertainty in the AERONET retrieved SSA is ~0.03 at AOD(440)~0.4 but that this uncertainty decreases for higher AOD levels (see Sinyuk et al. 2020: Fig 22 and Tables 14-17).
AR: Added a sentence to mention uncertainty in AERONET SSA decreases with increasing AOD.

RC: Line 667-668: This must be an error, or else you need to change the value of 0.05. Why only include the effect of surface reflectance in your estimate of retrieval uncertainty of SSA from satellite when there are several other significant sources of uncertainty such as AOD, aerosol layer height, aerosol size distribution, refractive index and satellite sensor calibration.
AR: Re-written the sentence to include an ensemble of error in our retrieved SSA due to all input variables.

RC: Line 671-673: However, please note in the text that the overlapping error bars comprise a wide range of +-0.08 which is a large fraction of the expected parameter space for aerosol single scattering albedo (~0.8 to 0.99).
AR: Removed the sentence that says… 'the errors in retrieved and AERONET SSA are consistent as shown with the overlapping error bars'.

RC: Lines 676-678: Please be aware that surface reflectance is a second order source of uncertainty for AERONET since the instrument is upward scanning, and the primary sources of uncertainty for AERONET retrievals of SSA are sky radiance calibration (which is independent of the direct sun cal), solar flux and AOD. For downward viewing satellite retrievals however the surface reflectance is a major source of uncertainty and therefore the way this sentence is written might possibly be quite misleading.
AR: Re-written the sentence(s) to include appropriate information from the complete error analysis.

RC: Lines 679-681: "For absorbing aerosols, the SSA differences observed here is likely a result of different surface reflectances data employed by the two data sets." No your statement here is not true at all. See my comment immediately above.
AR: Please see the above response.

RC: Lines 691-692: It should be noted that the uncertainty in SSA for AERONET is somewhat higher at 675 nm than at 440 nm for fine mode aerosol cases (for a given value of 440 nm AOD; see Sinyuk et al. (2020); Figure 22). This is due to the lower AOD at 675 nm therefore lower

absorption signal in the data. A similar increase in satellite uncertainty at the longer wavelength is inevitable for the same reason.
AR: Yes, we agree. As the absorption signal diminishes at higher wavelengths, surface reflectance contributes much of the upwelling radiances measured by the sensors. Therefore, high uncertainties of satellite retrieved SSA at longer wavelengths is inevitable particularly for weakly absorbing urban aerosols. While with AERONET the uncertainty in SSA primarily due to lower AODs at higher wavelengths.
Re-written the sentence for more clarity in the main text.

RC: Lines 703-705: It should be noted here in the text that the uncertainty in AERONET retrieved SSA decreases as AOD increases (Sunyuk et al. (2020) and that the same is true for the satellite retrievals since at high AOD the aerosol signal overwhelms the sources of uncertainty such as surface BRDF, calibration and AOD.
AR: Please see the above response. Re-written the sentence for more clarity in the main text.

RC: Lines 710-712: This statement is only partially true and needs to be revised. For dust aerosol at low Angstrom Exponent (AE) the AERONET retrieval imposes very weak constraint on the spectral variation of the imaginary refractive index since the AOD is high at all wavelengths and the absorption signal is therefore sufficient at all 4 retrieval wavelengths (440, 675, 9870 and 1020 nm). The sky radiances are fit well at all wavelengths for dust cases and therefore the retrieval is robust at all wavelengths. For fine mode dominated aerosol at high AE values however AERONET version 3 imposes a constraint on the spectral variation of imaginary refractive index. This constraint for high AE retrievals is based on the fact that black carbon exhibits minimal wavelength dependence in imaginary refractive index, plus the fact that for large AE the AOD at the longer wavelengths is quite low and therefore the aerosol absorption signal is insufficient for a robust retrieval at the long wavelengths.
AR: Revised the sentence(s) for more clarity.

RC: Lines 745-746: This statement is misleading as it suggests that the SSA is spectrally flat for the entire wavelength range. However in Table 3 the values for Mongu show significantly higher SSA at 466 nm than at 646 nm.
AR: In the summary of results, we are mentioning the shortest (340 nm) and longest (646 nm) wavelength SSA retrieved in this work. We have re-phrased the sentences for more clarity.

RC: Line 750-752: Again you are exhibiting a tendency to ignore the fact that in your Table 3 the SSA retrievals at 466 nm are significantly higher than at 646 nm for both the Alta Floresta and Missoula sites. Please explain the lack of consistency in your retrieval data versus your interpretations in the text of the paper.
AR: Please see the above response.

RC: Line 758-759: You call the Arabian Peninsula a biomass burning region? Please back up this interpretation with published references and some substantial analysis. Also regions such as eastern China and Northern India do have biomass burning in specific seasons however they are more strongly dominated by other emission sources for more months of the year and therefore are not considered biomass burning regions in the literature (except for specific ~1 month periods).

AR: Re-written the sentence to clearly mention the sites and region (Middle East) we are referring.

RC: Lines 761-762: Why did you only mention 466 nm SSA for this region in this Conclusions section? This section has been inconsistently written.
AR: Corrected for consistency in this section.

RC: Line 774-776: It is somewhat absurd to categorize the CUIABA site as representative of the urban aerosol type. The cases where the AOD at 440 nm exceed 0.4 at Cuiaba are almost always dominated by biomass burning smoke from cerrado vegetation plus some long-range transport of smoke from rain forest burning to the north (see Schafer et al., 2008)
AR: As per the aerosol categorization scheme employed using EAE and UVAI, we noted both carbonaceous and urban aerosols for the site at Cuiaba (see Table S1). We agree that Cuiaba is representative of biomass burning emissions. In the revised discussion (section 6.1) of biomass burning aerosols, Cuiaba is included.

RC: Line 779-781: Please provide some references to back up this interpretation, including discussion of the fuel types in most of Europe you are alluding to.
AR: Removed this sentence. Added appropriate aerosol sources for the sites in Europe.

RC: Line 795: This does not make a lot of sense and needs to be supported with additional references and analysis. The uncertainty of the retrievals at low AOD levels is inherently greater therefore this does not have much basis in rigorous analysis.
AR: Removed this sentence and eliminated the results of SSA at AODs lower than 0.4 in the manuscript.

RC: Lines 806-807: "Given the lack of aerosol absorption information at near-UV wavelengths in the existing AERONET record…" This is not a completely accurate statement. There is currently no AERONET product available on absorption at 380 nm but the sky radiances measurements have been made for years at 380 nm from many instruments in the global network. A retrieval that includes the 380 nm imaginary refractive index and SSA is currently under development by the AERONET project. This statement needs to be revised and expanded to reflect this additional information.
AR: We have now clearly mentioned about the availability of 380 nm sky radiance measurements and expected 380 nm SSA in the future upgrades of AERONET inversion product.

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