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

Reviewer – 3

RC: This work attempts to use the OMI-MODIS synergetic data and the measured and retrieved products from the AERONET measurements from different Earth regions. This work used UV-Vis spectral band measurements and retrieved the SSA separately for each bands using a LUT approach. Seasonal variability of the SSA in different regions worldwide is studied using the AERONET products and retrieved SSA and AAE from this work. In general, the attempt to quantify the aerosol absorption using the retrieved SSA in UV-Vis multi-year multi-dataset is interesting and useful for the scientific community. However, the methodology and the manuscript need to be improved before considering for publication.

We sincerely thank the reviewer(s) for providing detailed comments and suggestions wherever applicable that helped make substantial improvement of the manuscript. Major changes made in the revised manuscript are summarized as bullet points here.

- Performed sensitivity tests for all input variables used in our retrieval algorithm, including trace gaseous absorption (in the visible wavelengths) to provide an ensemble of theoretical uncertainty in the retrieved SSA.
- Aerosol type categorization from the combined the used of EAE derived from AERONET and near UV Aerosol Index (UVAI) from OMAERUV product.
- Clarification on the surface reflectance products used in the work and their uncertainty.
- Rewritten the entire section 5 (section 6 in revised version) with focus on the dominant aerosol type(s) prevailing over the worldwide regions.
- Replaced the line/bar plots with box plots wherever applicable to show the range of observations.
- Provided figures showing climatological seasonal aerosol size distributions used for developing LUTs.
- Corrected for appropriate references and other specific comments.

RC: AERONET measurement-based climatology is used for representing the particle size distribution in the retrievals. However, no discussion based on it was found in the manuscript. It would be ideal to provide the details of the PSD used in the retrievals. You could create similar plots like the figure 6,7,8, and 9; instead of SSA, you could plot the mean PSD distribution with the error bar as the SD (This can go to the supplemental material).

AR: The revised manuscript now includes figures showing the seasonal climatology of aerosol PSD as supplementary material.

RC: Authors should discuss the criterion for classifying AERONET stations into biomass burning, dust, Urban/Industrial, and mixed aerosol in detail. E.g., the Sao Paulo station classified as biomass burning is wrong since it is a megalopolis.

AR: We used aerosol typing information based on EAE derived from AERONET and UVAI obtained from OMAERUV product. Sao Paulo is not classified as biomass burning site. In the figures 6, 7, 8, and 9 we included the site names on the right upper corner with abbreviations D (dust), M (mixed), C (carbonaceous), U (urban) indicating the aerosol types that were averaged

to derive the regional spectral SSA. For the site at Sao Paulo, the only annotation used is 'U' indicating Urban type aerosols were observed over the site. In the revised manuscript, figures are now replaced, and the sites used in averaging are mentioned more clearly.

RC: Section 5 is not very clear. Please consider rewriting it to avoid ambiguity.

AR: In the revised manuscript, section 5 is re-written, and the results and discussion is limited to specific aerosol types.

RC: Consider moving Table 3 to supplemental material.

AR: Moved the table presenting seasonal SSA and AAE for all the sites to (now Table S1) supplemental material.

RC: A discussion on the correction of atmospheric gas absorption before the aerosols retrievals is needed.

AR: The RTM used in the work accounts for H₂O and O₃ absorption. To account for other unaccounted trace gases, we have now included sensitivity tests for the estimated optical depth of all trace gases that has absorption lines in the visible spectrum.

RC: L37: What models are authors discussing here? GCM? Global Earth System models? Please specify it.

AR: General circulation models (GCM).

RC: L55: Did you mean by SSA retrievals?. Because it is clear that there is a long term measurement of AOD at UV bands from AERONET stations.

AR: Yes, we mean lack of SSA retrievals at UV wavelengths in the existing AERONET inversion product.

RC: L71-73: Specifically mention these retrieval algorithms with citation. Are you specifically talking about MODIS operational algorithms?

AR: Added citations for the appropriate retrieval algorithms.

RC: L164: Provide geometry information. Like SZA, VZA, RAA used for simulating TOA observation for the GSFC site.

AR: Each site-specific LUT is developed for several fixed nodes of SZA, VZA, RAA. The figure 3 illustrates the simulated TOA radiances for selected geometry at SZA=20°, VZA=40°, and RAA=130°.

RC: L165-167: This is calculated for a particular satellite-sun geometry, and it can vary considerably in the analysis used in this study. How can you generalize this sensitivity of SSA and AOD to other satellite-sun geometries?

AR: Our purpose here is to demonstrate the well-known theoretical concept of LUT design and 2D-retreival domain of SSA, AOD typical of any satellite retrievals. Depending on the scattering angle, the magnitude of sensitivity for SSA and AOD might vary. In our retrievals, we use the associated satellite-sun geometry for each satellite-AERONET collocated observation.

RC: L177-180: Can you calculate the uncertainty in SSA due to the assumption made here?. It will be important, especially since retrieved SSA differs for regions with different spectral signatures and magnitude.

AR: As mentioned, we used a unified model to create non-spherical LUT of radiances for dust aerosols. This approach is similar to any typical satellite aerosol retrieval where a few models are assumed to retrieve global dust aerosols. In our work, we have not performed any sensitivity test on site- or region-specific dust aerosol particle sizes. This is a computationally extensive work and to avoid a much lengthy manuscript than what we already have we decided to explore this in our future studies.

RC: L184-186: Did you use the closest observation as collocated data?

AR: Re-written the sentence for clarity. We used the AERONET measurement closest in temporal domain to the satellite overpass.

RC: L203: Why the inversion is done independently? You could use the multiple wavelength information in minimization. What is the advantage of doing inversion independently over different spectral bands?. In the discussion section, you are using only the data points with retrievals for all bands. It makes sense to use all the information together to do a retrieval. Please specify the rationale behind not using this method.

AR: Since the UV and visible bands used in this work comes from OMI and MODIS sensors with different spatial and radiometric characteristics, we chose to derive the aerosol absorption independently. While the multi-spectral fit could be another approach to retrieve spectral SSA simultaneously, the minimization scheme often results in error in spectral fit at different wavelengths. Such technique seems to be more suitable for a multi-spectral and/or hyperspectral instruments carrying UV and VIS wavelengths. Nonetheless, an overall good agreement of the derived spectral aerosol absorption (AAE) in our technique with insitu data reported in the literature for different aerosol types demonstrating the effectiveness of the adopted approach.

RC: L212-214: This is not correct for the blue band. The Rayleigh signal will mainly dominate the signal in this band when the aerosol loading is low.

AR: This sentence is re-written to say that Rayleigh scattering gradually diminishes with increasing wavelength proportional to λ^{-4} .

RC: L270-271: How did you come up with these numbers? Provide references for this. It would be best to use the uncertainty specified by the surface albedo product you are using in the retrieval.

AR: For 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. The prescribed uncertainty in the derived layer height is expected to be within ± 1 km (Torres et al., 2013). The uncertainty in aerosol layer height mainly rises from very limited sampling of CALIOP overpasses (16-day repeat cycle).

RC: L282: This achievable accuracy depends on the accuracy of surface reflectance products used in this study. It should be calculated based on the accuracy mentioned for the surface product used and should differ based on surface type, and it will become dominant in the longer wavelengths.

AR: In the revised manuscript, all sensitivity are performed for the most relevant satellite-sun geometry and representative aerosol types observed at GSFC, Mongu, and Tamanrasset sites. To estimate the achievable accuracy in our SSA retrievals, we used a consistent ± 0.01 uncertainty in surface reflectance through UV and visible wavelengths. However, it should be noted that prescribed theoretical measurement-based uncertainty in the MAIAC MCD19A1 BRF product is much less, up to 0.002–0.003 at visible wavelengths.

RC: L325-327: I can't see a plot for the DJF season. Is this a typo? AR: Yes, it is a typo. We corrected this as JJA.

RC: L366-367: This can be verified using the plots of PSD from AERONET retrievals. AR: We presented the figures showing PSDs as supplementary material. It is evident from the aerosol PSD at Ilorin that both fine-mode and coarse-mode particles are noted during DJF.

RC: L369-370: Is this just the author's opinion? What are the pieces of evidence for this? AR: Included appropriate reference showing similar result.

RC: L395-396: For the case of JJA, the SSA is increasing with the wavelength for the region 340-388 nm. It is contradicting to the sentence in these lines. AR: Re-written the sentence for clarity.

RC: L405: Typical urban spectrum based on what work? Cite the literature! AR: In section 6 (revised version), we discussed the spectral behavior of each representative aerosol type with references before presenting any results.

RC: L422-423: This has to be verified using PSD.

AR: The average aerosol PSD obtained for the European sites show both fine-mode and coarsemode particles with nearly similar volume concentrations. The $\alpha_{440-870}$ for these aerosols fall in the intermediate-mode category as defined in our work as 0.2 - 1.2.

RC: L518-521: The SSA difference for those two regions can be due to the error from surface reflectance estimation.

AR: Re-written this sentence with references that also shows differences in SSA obtained for dust aerosols over Sahara and Sahel.

RC: L521-522: This is just another hypothesis; there is no proof here.

AR: We have re-written this sentence with references that also shows differences in SSA obtained for dust aerosols over Sahara and Sahel.

RC: L555: What kind of interpolation?

AR: AERONET SSA at 440 and 675 nm are used in linear interpolation to derive SSA for MODIS wavelengths at 466 and 646 nm.

RC: L560: In figure 13, why there is no STD for the UV wavelengths?

AR: There is no STD for UV wavelengths, since we are showing differences in SSA with AERONET at visible wavelengths. In the revised manuscript, we replaced this line plot with box plot to show the lower and upper quartile range of observations.

RC: L561-563: Show the SSA as a box plot with error bars. It will give us an idea of the spread of the SSA values for the averaging AERONET station.

AR: Revised figure now shows the data as box plot with lower and upper quartile range of observations.

RC: L576: Move the annotations to the lower right corner of the plots in Figure 14. AR: Moved the placement of annotations for visual clarity.

RC: L601: You could cite Dubovik et al., 2006 to describe the smoothness parameter imposed in the retrieval.

AR: Included the appropriate reference for this sentence.

RC: L601-603: Another difference is the use of multi-angular- multispectral information in the AERONET retrieval, Whereas the work presented here used the PSD and real refractive index from those retrieval and basically, the imaginary part of the refractive index is varied to retrieve the SSA.

AR: Yes, thanks for pointing out this difference for us.

RC: L616-618: Define the range of parameters used for this sensitivity study.

AR: Re-written this sentence to include complete set of input variables used in the sensitivity tests and provided an ensemble of error estimated in the retrieved SSA.

RC: L661-662: These two AERONET stations is in the biomass burning aerosols category. Then why you have it here in Urban?

AR: In the figure 6a, we annotated the sites with D, M, C, and U indicating the aerosol types. In our sample for CUIABA both carbonaceous and urban aerosols were observed, while for Sau Paulo only urban aerosols were found. As mentioned earlier, the aerosol-typing scheme employed here uses combined EAE derived from AERONET and near UV Aerosol Index (UVAI) derived from OMAERUV product. The revised manuscript provides new figures clearly indicating the sites used the regional average of spectral SSA.

RC: L687: Specify that no SSA and IRI retrievals available at the moment.

AR: We have included clear statements now mentioning the wavelengths in the existing inversion product and the possibility of AERONET SSA and IRI at 380 nm in the future.

Technical corrections RC: L40-41: Is it a 50% decrease? AR: Corrected as 'decrease'.

RC: L59: Citation required for this statement. AR: Included citation.

RC: L72: 'observations in the visible assume'. It should be 'visible spectrum' instead of just 'visible'.

AR: Corrected as 'visible spectrum'.

RC: L124: In Table 1 it is mentioned that version 2 is used. Which one is correct? AR: For developing LUT radiances we used version 2 level 2 aerosol particle sizes. Other than that elsewhere (i.e., AOD inputs for the retrievals and SSA comparison) in our work version 3 level 2 AERONET products are used.

RC: L198-199: Did you mean an exponential distribution?. Because the peak is on the surface. AR: Yes, exponential distribution with a peak on the surface.

RC: L290: Is it τ440 ≥0.2?

AR: Yes, it should be $\tau_{440} \ge 0.2$. However, the revised manuscript and supplementary materials now includes only observations with $\tau_{440} \ge 0.4$.

RC: L305: The values given for SSA are mean; specify it explicitly with the SD as uncertainty. AR: We have now provided the mean SSA and SD in the main text discussions.

RC: L441: In figure 9b, instead of 'northeastern china', it is mentioned as 'eastern china.' AR: Corrected the region as 'northeastern China'.

References

Lyapustin, A., Wang, Y., Korkin, S. and Huang, D.: MODIS Collection 6 MAIAC algorithm, Atmos. Meas. Tech., 11(10), 5741–5765, doi:10.5194/amt-11-5741-2018, 2018.

Torres, O., Ahn, C. and Chen, Z.: Improvements to the OMI near-UV aerosol algorithm using A-train CALIOP and AIRS observations, Atmos. Meas. Tech., 6(11), 3257–3270, doi:10.5194/amt-6-3257-2013, 2013.

Torres, O., Bhartia, P. K., Jethva, H. and Ahn, C.: Impact of the Ozone Monitoring Instrument row anomaly on the long-term record of aerosol products, Atmos. Meas. Tech., 11(5), 2701–2715, doi:10.5194/amt-11-2701-2018, 2018.