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

Reviewer – 2

RC: This manuscript presents a method for deriving aerosol absorption from a combination of satellite and ground-based remote sensing measurements. The single scattering albedo is derived in five wavelengths in the UV-Visible range. The method can be applied over locations with co-located satellite and ground-based measurements. The method adds UV information on top of the existing data from AERONET. The method is applied to a data record over more than 100 sites globally for the period 2005 – 2016. Results are discussed per region.

Overall, I think this is interesting work, which has a lot of potential. However, the way that it is presented can be much improved. I recommend splitting this manuscript in two parts, where part I describes the method, sensitivity study, case studies and validation, and the part II describes global and regional results. Part I would fit for AMT, whereas part II would better fit in ACP or a similar journal. Part I should answer questions like, what is the added value of this method with respect to the standard AERONET retrievals of SSA? How can the retrieved spectral behavior of the SSA be explained by the expected refractive index? That is why I encourage the authors to withdraw the current manuscript and resubmit it in two improved parts. Because of this recommendation I will not provide detailed textual comments on the current manuscript, but rather indicate where the work needs further improvements.

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: Section 3: Before section 3.1 a text needs to be added that introduces the methods physical background. E.g. what determines the radiation measured at ground-based and satellite level. A diagram would be useful for this.
AR: Added description of the physical basis (section 1) of the satellite remote sensing of aerosols.

RC: Section 3.1: The selected wavelengths in the visible are affected by NO2 and ozone absorption. However, these absorptions are apparently not part of the LUT design. These should be included, or the authors should justify why these absorptions can be neglected. In the end, the method uses reflectances from the satellite instruments and derives the SSA using the LUT. Therefore, the SSA is the depended parameter for the method. For this reason, the axes of Figure 3 should be switched (x-axis reflectance, y-axis SSA). This will immediately visualize the problem for low AOD, where the method will be very noisy.
AR: Revised manuscript now includes estimated error incurred due to unaccounted trace gases in the LUT radiances. Revised the figure 3 as suggested.

RC: Section 3.3: It is unclear where the surface pressure information is coming from.
AR: Included surface pressure information in section 3.3.3. For OMI the surface pressure is directly adopted from OMAERUV product, while for MODIS the terrain height available in the aerosol product is used to convert it to surface pressure as shown in the below equation.

$$P(z) = P_o \exp\left(-\frac{z}{H}\right)$$

Where, P_o is the sea level pressure 1013.25 hPa, and H is scale height of the atmosphere 7.5 km.

RC: Section 4: The sensitivity analysis is incomplete. The approach to only perform a sensitivity analysis for parameters, which are controlled in the retrieval, is clearly not acceptable and also not true, because the real part of the refractive index and other aerosol model parameters are also selected as part of the algorithm. So, all identified parameters should be included in the sensitivity analysis, along with the surface pressure, signal to noise of the instruments, the (tropospheric) ozone column and the NO2 column. This should be presented in graphical way to convince the reader that the method is sound.
AR: Revised manuscript now includes sensitivity tests quantifying the error in SSA due to perturbation in all input variables.

RC: While the analysis of the GSFC method is good, I am left with a number of questions. First of all, the SSA should significantly lower values at 646 nm compared to the other wavelengths. Is this realistic. Is this in line with our knowledge about the refractive index in the visible? Is it possible that this is related to ozone absorption in the visible?
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 changed the x-axis to show AOD400. "The mean aerosol SSA retrieved at the GSFC site for observations with $\tau_{440} > 0.4$ at 340, 354, 388, 466 and 646 nm are 0.94, 0.95, 0.95, 0.94 and 0.93, respectively. These results agrees well with the values reported for GSFC site using AERONET products at 440 and 675 nm as 0.96 and 0.95, respectively (Giles et al., 2012)."

RC: I propose that on top of the GSFC analysis, the authors present 3-6 cases studies of single retrievals over different sites, where for a given day for which both the AERONET SSA and the combined SSA retrievals are available. These cases should cover both good and bad comparisons and discuss the reasons for these results. This gives an opportunity to demonstrate the added value of the satellite method.
AR: We thank the reviewer for encouraging to present more case studies of retrievals over different sites. However, we presented at least 12 sites (4 sites for each representative aerosol types) including GSFC in comparison of SSA section. These sites cover both good and not so

good comparisons showing mean SSA differences up to 0.05. We replaced the line plot to show the central lower and upper quartile range of observations (figure 8 in the revised version). Given the already lengthy manuscript, we decided to use only one site (GSFC) in the section 3.3 to demonstrate our results.

RC: Section 7: Logically, the next section would be the validation (currently section 7). The authors have chosen to compare only the 466 and 646 nm SSA to AERONET. Also comparison between 388 nm retrievals and 440 nm AERONET shall be included. Although the spectral distance is larger than for 466 nm (MODIS), it is the best way of also including the OMI retrievals in the validation. The validation data should also be split by AOD bin. In this way I hope that some correlation can be demonstrated for the medium and high AOD values. Alternatively, times series of data sets over sites with large variability in SSA could be presented to convince the reader that the retrievals add value wrt to the results from AERONET. The current Figures 14 and 15 (top plots) are not very convincing, however the representation of Figure 15 (bottom) is much better.
AR: We have moved the validation section to top, immediately after the error analysis. SSA comparison figures now include data presented as difference in SSA as a function of τ_{440} for individual aerosol types (carbonaceous, dust, and urban).

RC: Section 5: In section 5 regional results are presented. There are different ways of computing the average SSA. If I understand it correctly, the unweighted average SSA is presented. Alternatively, given the dependence of the accuracy on the AOD, the AOD could be used as weights. This would be equivalent to computing the mean SSA as (1-mean(AAOD)/mean(AOD). Also, mean and standard deviations can be significantly affected by outliers, whereas the median and percentiles are more robust statistics. How would the presented results be affected if other methods of computing statistics are used?
AR: Yes, we presented the unweighted average SSA for all observations with $\tau_{440} > 0.4$ in the regional results (section 6 in revised version). However, examination of the median SSA values (not presented in the manuscript) does not show any significant differences than the unweighted mean values reported here.

RC: The results are presented per region. However, given the poor spatial sampling the statistics will not be representative for the whole region. It is therefore questionable if this analysis is useful at all. For example, there is a huge difference between aerosols on the Californian coast and those of continental Canada, however they are in the region. The same is true Mediterranean sites and sites in Northern Europe as well as other regions. The authors should rethink how these data can be best presented, beyond the current split in regions. I suggest starting with some global maps where the data is plotted per season and aerosol type. Maybe as a circle of which a quarter is used per season or something similar.
AR: We agree that given the poor spatial sampling the regional averages presented here may not be representative for the entire regions. We mentioned it in the main text clearly. The revised manuscript uses appropriate region names than previously mentioned to avoid ambiguity.

RC: Section 5 is very hard to read, as it mixes observations with speculations. Also results from large regions are discussed in terms of very local phenomena. Here a clear choice should be made by authors to either discuss the global distribution of the SSA, or to dive into the details of

one or more regions. Now the scope is somewhere in between and that doesn't work for me. Furthermore, it should be clearly identified when the other claim that the data prove something, or when they speculate about possible explanations.
AR: We have rewritten the entire section 5 for more clarity.

RC: In many cases in Figure 6/7/8/9 a significantly lower SSA at 648 nm is reported as compared to the other wavelengths (e.g. 6a/b, 8 a/b/c, 9 b/c). Do the authors have an explanation of this, in terms of the spectral behavior of the refractive index? Is this reported in other studies? I am not convinced that this is not caused by measurement errors.
AR: It is well known that for other than dust, spectral SSA of aerosols will decrease with increasing wavelength from UV to Visible spectral range. For the results presented in regional analysis the curvature of spectral SSA is consistent with the known behavior. We included comparisons of SSA and derived AAE with insitu measurements and AERONET analysis over the corresponding sites available in the literature. Given the high uncertainties of retrieved SSA in visible wavelengths, the retrieved SSA and AAE compare very well demonstrating the effectiveness of our technique.

RC: Section 6: I am not convinced by the analysis based on the mean AAE. Overall, satellite retrievals of the AE are difficult and the AAE is much more difficult. Also note that AAE is a combination of the spectral behavior of the AOD and that of (1-SSA). Before concluding jumping to conclusions on the AAE, the authors should first provide that there is any value in these mean AAE results.
AR: We agree, satellite-based retrieval of AE and AAE are hard to quantify. In the revised manuscript, we provide sensitivity test by varying the retrieved SSA by the estimated uncertainty. We clearly mentioned in the text that AAE values provided here are for qualitative or informational purposes, as small changes in SSA would incur wide range of AAE.

RC: Tables 3. I propose remove this table to the supplemental material and make it available as complete data set (e.g. HDF5 file(s) or excel files(s)) for all the sites, containing both individual retrievals and statistics.
AR: Table 3 is now moved to supplemental section. We are still in the process of exploring the complete retrieved data set and will look for the opportunity to make it publicly available in a suitable format.

RC: Figure 1. Move this to supplemental material.
AR: We chose to keep this figure in the main text to show all the sites used in our SSA retrieval procedure.

RC: Figure 10-12, replace the bars by violin plots or box-whisker plots.
AR: Revised manuscript has these figures now shown as boxplots with lower and upper quartile range of observations.

RC: Figure 13, replace the plots by box and whisker plots using the spread of the points instead of the assumed uncertainty.
AR: Replaced this figure with boxplots with lower and upper quartile range of observations.

References

Giles, D. M., Holben, B. N., Eck, T. F., Sinyuk, A., Smirnov, A., Slutsker, I., Dickerson, R. R., Thompson, A. M. and Schafer, J. S.: An analysis of AERONET aerosol absorption properties and classifications representative of aerosol source regions, J. Geophys. Res. Atmos., 117(D17203), 1–16, doi:10.1029/2012JD018127, 2012.