Retrieving UV-VIS Spectral Single-scattering Albedo of Absorbing Aerosols above Clouds from Synergy of ORACLES Airborne and A-train Sensors

Jethva et al.

Response to the Anonymous Referee # 2

We thank and appreciate the anonymous reviewer for providing constructive and valuable comments on our manuscript.

Before responding to each of the comments provided by the reviewer, we want to let both reviewers and the Editor know about the major upgrade we applied to the scientific content presented in our paper. The need to apply a major revision stems from the comments made by referee # 1 regarding the cloud model properties. Along the process of responding to the comments, it was realized that the cloud effective radius assumed in the C1 cloud model adopted in the present work may not be an optimal representation of the stratocumulus clouds observed in the Southeastern Atlantic Ocean during the ORACLES operation period.

The figure shown below displays the histogram of the cloud effective radius (CRE) data derived from the standard Aqua/MODIS MYD06 cloud product over the Southeastern Atlantic Ocean from August through October 2016 to 2018. The CRE histograms for the months of August to October reveal a distinct peak around 11-12 μm with a wide distribution ranging from 6 μm to 26 μm, albeit 70%-80% of the data points are found to be within 6-18 μm. Meyer et al. (2015) have shown that the presence of partially absorbing carbonaceous smoke aerosols over clouds in this region has only a marginal impact on the retrieved CRE.
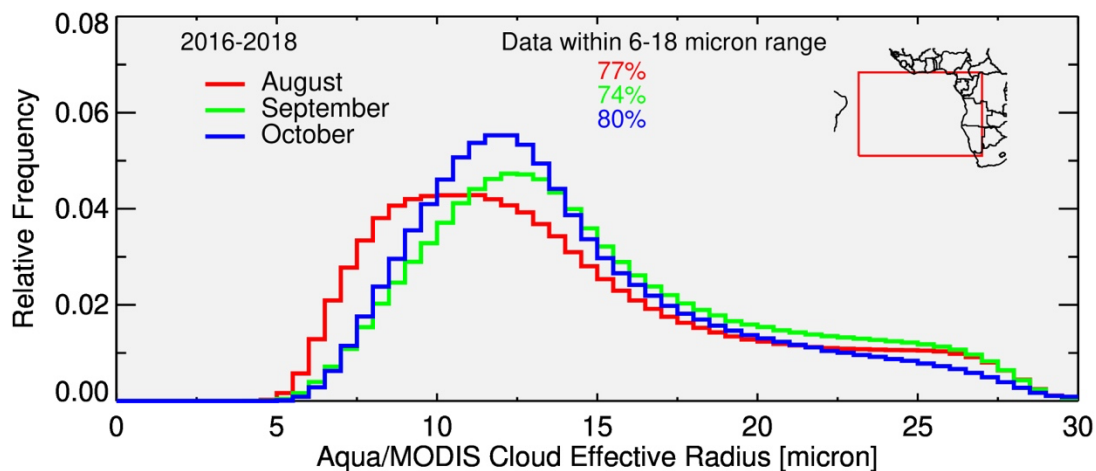

Figure 1 Histograms of the Aqua/MODIS cloud effective radius retrievals over the Southeastern Atlantic for the ORACLES period of August through October 2016-2018.

The C1 cloud model adopted in our work assumes the liquid droplet size distribution that follows the modified Gamma distribution with α, β, and γ parameters of 6.0, 1.5, and 1.0, respectively. The CRE value, when calculated using these parameters, was 6 μm, which is an underestimation compared to the MODIS CRE histograms discussed above.

Based on the MODIS CRE histograms corresponding to the region and period of interest to our study, therefore, we have revised the cloud model with an optimal CRE value of 12 μm. This is achieved by changing the α parameter from 6.0 to 15.0 and keeping the β and γ parameters unaltered.

The entire analysis presented in the originally submitted manuscript was reperformed with the revised cloud model that required recreating the aerosol-cloud LUTs for the above-cloud SSA retrieval and its sensitivity analysis. The following are the major changes we noticed in the results.

1. The derived above-cloud SSAs are now lower in magnitudes by up to 0.01 and 0.02 at the near-UV and VIS-NIR wavelengths, respectively.
2. Most importantly, the comparison of the satellite-retrieved SSA against ORACLES in situ measurements, 4STAR inversions, and ground-based AERONET inversions improved noticeably, where the overestimation seen in the satellite retrievals is now either eliminated or largely reduced.
3. The results of the sensitivity analysis tabulated in Tables 3 and 4 show marginal change without its interpretation discussed in the original version of the manuscript.
4. The revised paper now also includes the uncertainty estimates in the retrieved aerosol-corrected COD resulting from perturbing the assumed aerosol and cloud parameters. These results are tabulated in Tables 5 and 6.

All relevant figures and tables are updated with the revised calculations and results.

Referee comment **RC in RED**
Author's response **AR in BLACK**

General comments from Referee # 2

"Retrieving UV-VIS Spectral Single-scattering Albedo of Absorbing Aerosols above Clouds from Synergy of ORACLES Airborne and A-train Sensors" by Hiren Jethva et al.

This paper introduces a synergy algorithm to improve the retrieval of SSA of aerosols over clouds, which is dearly needed to improve estimates of aerosol direct effects and aerosol forcings. The method is based on a 'color ratio' approach which relies on the spectral absorption of aerosols in the UV and visible, which is true mostly for smoke. Using two spectral measurements, two pieces of information can be retrieved, like AOD and COD when assuming the spectral properties of the aerosols. In the current paper, a method is proposed to instead constrain the AOD above the cloud and retrieve the COD and SSA of the aerosols, which can be done if independent above-cloud AOD is available, in this case using airborne measurements from three ORACLES campaigns over the tropical Atlantic. This area is interesting for the smoke over persisting stratocumulus, providing an obvious focus for the application provided analyses.

The presented approach is interesting since SSA retrievals, especially over clouds, are scarce. The application of the ORACLES aircraft campaign data is an interesting approach to test the satellite retrievals with a proper constraint on the aerosol properties, which is almost always missing. However, there are some major issues related to the presentation of the work.

First, the introduction is quite extensive, but it reads like an advertisement for the color ratio approach, while many pioneering work has been neglected.

Some examples:

Hsu et al. (2003) did not perform actual retrievals of aerosol-cloud properties of the elevated smoke layer above the cloud deck. Instead, they showed the impact of partially absorbing smoke layer overlying clouds on the satellite measurements of Reflectivity, spectral reflectance, and reflected solar radiation from clouds. Since Hsu et al. (2003) paper was about the direct effects of aerosols above clouds, we have added the reference following the statement, "Unlike the cloud-free conditions in which aerosols generally produce a cooling effect on climate, the presence of elevated layers of absorbing aerosols over clouds can potentially exert a large positive forcing through enhanced atmospheric heating resulting from aerosol-cloud radiative interactions (Hsu et al., 2003; Keil and Haywood, 2003; Chand et al., 2009; de Graaf et al., 2012; Zhang et al., 2016; Kacenelenbogen et al., 2019)".

Peers et al. (2019) demonstrates the retrieval capability of aerosols above clouds (AOD, COD, and CRE) from MSG/SEVIRI. Therefore, we've mentioned this reference in the statement "These techniques have shown the potential to retrieve ACAOD using measurements from different A-train sensors, including CALIOP/CALIPSO (Hu et al., 2007; Chand et al., 2008), Aura/OMI (Torres et al., 2012), Aqua/MODIS (Jethva et al., 2013; Meyer et al., 2015), Parasol/POLDER (Waquet et al., 2009), and MSG/SEVIRI (Peers et al., 2019)."

Meyer et al. (2015) also showed impact of absorbing aerosols above clouds on the retrieved COD and CRE. We've added both Haywood, et al. (2004) and Meyer et al. (2015) papers here.

The physical basis of the color ratio method explained in our previous papers corresponds to the simultaneous retrievals of ACAOD and COD, by assuming a fixed aerosol model, including SSA. In this paper, since we're proposing to retrieve SSA above clouds by constraining the AOD as an input, a separate figure demonstrating the sensitivity of TOA reflectance and ratio is needed. The six-panel Figure 1 shows the distinct sensitivity of color ratio and reflectance to the varying imaginary part of the refractive index or SSA (since size distribution is fixed) and COD for three different values of input ACAOD. Therefore, we will retain Figure 1 in its original form.

Agreed. The sentence is modified as "In other words, this kind of situation produces a strong 'color ratio' effect, which can be seen in the TOA measurements made by satellite sensors such as OMI (Torres et al., 2012), MODIS (Jethva et al., 2013), and OMI-MODIS combined (De Graaf, et al., 2019).

l 530. "The lofted layers of absorbing aerosols over the clouds attenuate light reflected by the cloud top through scattering and absorption. This effect reduces cloud-reflected upwelling UV (Torres et al., 2012) and VIS-NIR radiation (Jethva et al., 2013; Meyer et al., 2015),"

Under the pretence of "benchmarking" ones own RTM:

l. 366 The model results have been benchmarked against the results published in the literature, as well as used in the operational ACAOD product of OMI (Jethva et al., 2018), research-level ACAOD retrievals from MODIS (Jethva et al., 2013, 2016), DSCOVR-EPIC (Ahn et al., 2021), and S5p-TROPOMI (Torres et al., 2020).
Here, we want to say that the VLIDORT model results were compared against the RT simulations from benchmark RT codes, such as TOMRAD and the ARIZONA code used internally within the trace gas-aerosol groups in NASA Goddard. The same VLIDORT code was further upgraded for the joint aerosol-cloud simulations and used to create look-up tables for the above-cloud AOD retrieval from OMI, EPIC, and TROPOMI. The validation of the ACAOD product from these sensors against the ORACLES airborne direct measurements revealed a good level of agreement, suggesting the accuracy and reliability of the VLIDORT RT code.

We have rephrased this sentence for better clarity.

"The VLIDORT model results were compared against the published literature and the RT simulations from benchmark in-house RT codes used within the trace gases and aerosol groups at NASA Goddard. The VLIDORT code was further upgraded for the joint aerosol-cloud simulations and used to create look-up tables for the above-cloud AOD retrieval from Aura/OMI, DSCVOR-EPIC, and S5p/TROPOMI sensors. The validation of the ACAOD product from these sensors against the ORACLES airborne direct measurements revealed a satisfactory agreement within the expected uncertainties (Jethva et al., 2018; Ahn et al., 2021; Torres et al., 2020), thereby establishing the reliability of the VLIDORT RT code in the present application."

Repetition of references:

l. 85 These techniques have shown the potential to retrieve ACAOD using measurements from different A-train sensors, including (...), Aura/OMI (Torres et al., 2012), Aqua/MODIS (Jethva et al., 2013; ....),

l 102. This work builds upon the 'color ratio' (CR) technique previously applied to OMI (Torres et al., 2012; Jethva et al., 2018) and MODIS (Jethva et al., 2013, 2016) sensors.
We don't see any problem with this sentence or self-citation, as it is true that the technique proposed in the current paper is an extension of the CR algorithm that our group has developed and applied previously to the OMI and MODIS observations for retrieving ACAOD.

l. 122. In other words, this kind of situation produces a strong 'color ratio' effect, which can be seen in the TOA measurements made by satellite sensors such as OMI (Torres et al., 2012) and MODIS (Jethva et al., 2013).
Addressed in the one of the previous comments.

l. 364. We employ the VLIDORT code (Spurr, 2006) (...) - This should be merged with the info on l. 130.
We retain this sentence at its original place.

l. 718 - l.724: For instance, Jethva et al. (2018), through theoretical simulations, quantified (...) - Remove from the summary or merge with results in 5.4.

The sentence is now shortened and rephrased as "A perturbation of ±0.03 in the assumed aerosol SSA can lead to the uncertainty in the retrieved ACAOD in the range -20% to +50% at the UV and VIS wavelengths, the variation of which depends on the magnitudes of both ACAOD and COD."

Second, the manuscript lacks a comparison with existing work. Section 6 is interesting, if it would be compared to what we already know. This could easily be a simulation of the change in retrieved COD as a function of the aerosol absorption (in whatever form, SSA or AEE), which should not be too difficult with the RTM and LUTS already available. Alternatively, and more interesting would be to compare the actual measurements of the COD retrievals with and without ORACLES constraint aerosol above the cloud to other estimates of the change in COD, like e.g. in Haywood, et al (2004) or Peers, et al (2019). Now we learn nothing new. The main conclusion of the section is this, which should be put in perspective:

AR: Figure 11 (a) precisely demonstrates the effect of accounting (or not) for the presence of absorbing aerosols in the cloud optical depth retrievals. The revised manuscript includes the following description on the comparison of COD retrieved with and without using ORACLES-constrained ACAODs.

"*The aerosol-corrected CODs are found to be higher compared to apparent CODs (not corrected for aerosols) by an average (standard deviation) value of 26% (±20%) and 9% (±7%) at 388 nm and 860 nm, respectively. In other words, if not accounted for the presence of absorbing aerosols above the clouds, the retrieved COD turns out to be about -18% (±14%) and -8% (±6%) biased low compared to the aerosol-corrected CODs at UV and NIR wavelengths, respectively. The linear regression fits show positive slopes of 1.56 and 1.21 at 388 nm and 470 nm, respectively, with a negative intercept of ~1-1.5, which points to the inherent uncertainties involved in the proposed inversion technique at lower COD values.*"

Furthermore, a parameterization of the differences in CODs as a function of the above-cloud AAOD shown in Figure 11 (b) and (c) brings relatively new information that the former quantity is a bi-function of both the true COD underneath the aerosol layer and AAOD. These results are important for a more accurate estimation of the radiative effects of aerosols above clouds.

RC: l. 556 - 560. "These results are significant and further signify the importance of aerosol absorption above clouds in the UV to VIS-NIR spectral region in at least two ways. First, aerosol absorption above clouds, if not accounted for in the remote sensing inversion, can potentially introduce a negative bias in the retrieved cloud optical depth retrieval, whose magnitude depends on the strength of aerosol absorption (AAOD) and cloud brightness (COD) underneath the aerosol layer. " It would be nice to see how significant actually the results are, by comparing it with the expectations and results from earlier studies.

AR: The negative bias in the retrieved cloud optical depth, resulting from not accounting for the presence of overlaying aerosol layers, has been shown earlier in our previous papers and by other researchers. The results presented in this paper, i.e., Figure 11 (a), are a reaffirmation of the previous findings, using the airborne-satellite synergy. The impact of these results on the estimation of aerosol direct radiative effects requires a separate study, which is out of the scope of the presented work.

RC: Also, the sensitivity section is interesting and the amount of work is appreciated, it provides the necessary feel of the accuracy of the proposed method, and AMT is the right place for this kind of information. However, the provided tables of increased or decreased numbers of quantities are tediously repeated from the table into the text, and not compared to anything. It would quite interesting to have a few sensitivity studies from the RTM be compared to the retrievals. Then we

could actually see if what is expected is also being found from the satellite measurements when aerosol properties are properly and sufficiently constrained from the aircraft campaigns, or that still more information is needed.

AR: The approach we adopted to quantify the theoretical uncertainty presented in our paper combined both the radiative transfer simulations or aerosol-cloud LUT and the perturbation of actual measurements of ACAOD, ALH, AAE, and PSD. For instance, perturbing ACAOD and ALH did not require the generation of additional LUT. Instead, both quantities varied within the expected range of uncertainties (due to errors in measurements and/or airborne-satellite collocation) to calculate the change in the retrieved SSA. On the other hand, the sensitivity of the SSA retrievals to the change in AAE, PSD, NO_2, and cloud effective radius did require additional RTM and LUTs. Furthermore, the entire sensitivity analysis was applied to the actual airborne-satellite dataset of HSRL-2-OMI-MODIS synergy for the ORACLES August-September 2016 to quantify the realistic error matrix in the retrieved above-cloud SSA and cloud optical depth (the latter is added in the revision).

We don't think sensitivity studies purely based on RTM would add any additional information or improve the error matrix reported in our paper.

RC: Last, the manuscript lacks a proper motivation of the proposed synergy method. Obviously, the ORACLES data can only be used to test a proper constraint of the aerosol during the flights. CALIOP is mentioned as a replacement of the ACAOD measurements, noticing that (l776.) "CALOP [sic], OMI, and MODIS sensors fly in formation and make measurements within a few minutes of time difference.", in the present tense. The Calipso mission has ended and OMI is hardly producing useful measurements any more. Is the purpose to derive a SSA-over-cloud climatology from the past 10-15 years? Or can this also be applied to existing and future missions?

AR: We believe the motivation for developing the proposed synergy method is adequately discussed in the Introduction section. The above-cloud AOD retrieved from the passive sensors (such as OMI & MODIS) using multispectral algorithms is sensitive to the choice of aerosol model, as shown in our previous publications as well as by others (e.g., Meyer et al., 2015). Of various assumptions made in the model, the single-scattering albedo, followed by the spectral dependence of absorption (or AAE), is known to be the largest source of uncertainty in the retrieved ACAOD. These uncertainties, if not addressed, can proportionately result in errors in above-cloud aerosol radiative effects. Radiative transfer simulations show that the top-of-atmosphere measurements are sensitive to columnar ACAOD, SSA, and COD. Therefore, constraining the ACAOD in the algorithm allows two-parameter retrievals of SSA and COD. To test our hypothesis and the proposed algorithm, we used direct airborne measurements of ACAOD collected during the ORACLES campaign synergized with the OMI and MODIS observations over the Southeastern Atlantic Ocean.

A successful application of the synergy algorithm to the ORACLES and OMI-MODIS observations opens a new opportunity to extend the algorithm to CALIOP-OMI-MODIS synergy. Yes, we acknowledge that the CALIPSO mission has ended, and its light was turned off sometime this summer. OMI on Aura is also heading towards the end of its life. So, the intended application

of the proposed algorithm is to produce the past record of SSA above clouds from the coincident and overlapped period, i.e., 2006-2020.

RC: Or can this also be applied to existing and future missions?
AR: The proposed algorithm relies on accurate measurements of ACAOD for the retrieval of above-cloud aerosol SSA. The ACAOD algorithms applied to the existing passive sensors, such as OMI, MODIS, and POLDER, are dependent on the optical-microphysical models of aerosols. These retrievals carry errors due to the uncertainties in the assumed aerosol model. We're unaware of any existing satellite sensor that provides direct measurements of AOD above the cloud. Regarding the future missions, an HSLR-2 like active lidar sensor onboard satellite mission would provide the desired direct measurements of ACAOD for the proposed SSA retrievals.

I feel there is an increased interest in aerosol-cloud-radiation interaction and upcoming mission focus exactly on this topic, like 3MI/Sentinel-5 and EarthCare. The results presented here would be interesting for those missions as well and deserve a discussion, especially in a section named FUTURE IMPLICATIONS.
We have added a brief discussion on the applicability of the proposed synergy method to the future AOS and EarthCARE missions in section 8.1: Application of the Proposed Synergy Method to Existing and Future Missions.

Some textual issues are listed below and should be addressed, but more importantly some terms and phrases must be revisited:

(1)

l 639. "A strong relative spectral dependence of 20% in the imaginary part of the refractive index between 354 nm and 388 nm, resulting in AAE of 2.45-2.60 "

This should be rephrased in proper physical terms. It is unclear what is meant, but from the context one can expect that the spectral variation of the refractive index is meant, which cannot be "strong". It also does not result in AAE of 2.45, it is expressed in AAE.
We have reworded the sentence for clarity: "*The smoke particles are assumed to be, so-called 'colored aerosols', representing spectrally varying imaginary part of the refractive index, in which the latter is assumed to be 20% higher at 354 nm than that at 388 nm (see Table 2). This spectral dependence in the imaginary part of the refractive index translated into AAE in the range 2.45-2.60, which is expected to adequately represent the organic content of the carbonaceous smoke from biomass burning.*"

Similar arguments hold for:

l. 753. "reduced spectral dependence of absorption". Please, state the reduction in AEE (if this is meant).
Yes, we meant the reduced spectral absorption as reduction in AAE. The sentence is now rephrased as "*Changing the near-UV aerosol model with reduced spectral dependence of absorption or*

relatively lower AAE brought the observed RMSD down from 0.05 to ~0.03—an improvement of 0.02 in SSA."

l. 757. "stronger absorption strength" Please, rephrase.
The sentence is now rephrased as *"The satellite retrievals, ORACLES in situ measurements, and ground-based inversion from AERONET all show increased absorption (lower SSA) in August, relatively weaker absorption in September, and even lower absorption (higher SSA) in October".*

(2)
Replace all instances of 'spacetime' with spatiotemporal (or something better).
Both 'spacetime' and 'spatiotemporal' collocation convey the same meaning. However, we have adopted reviewer's suggestion to use 'spatiotemporal' in the revision.

Minor issues:
l. 64-67: A better (or additional) reference of the critical cloud fraction is Chand, et al (2009).
Chand et al. (2009) varied ACAOD, Angstrom Exponent, and asymmetry parameter, in their RT simulations, but kept SSA fixed as 0.85±0.02, whereas Eswaran et al. (2015) showed the sensitivity of CCF to varying SSA. We find both studies complementary to each other. We have added the following sentence referring to the work of Chand et al. (2009).

"*Along the same line, Chand et al. (2009) estimated the critical cloud fraction to be 0.4 for the Southeastern Atlantic Ocean, and further stated that it is strongly sensitive to the amount of solar radiation absorbed by aerosols, which in turn depends on the columnar aerosol loading and absorption capacity (SSA) of aerosols, and the albedo of the underlying clouds.*"

l. 181-184. This information seems out of place for the current paper.
The information given here simply states that the 4STAR sun photometer was successfully operated during the past field campaigns and provided invaluable optical measurements of aerosols and trace gases.

Section 5.1 should be merged with section 3.1.1. and repetition of information removed.
Sections 3.1.1, 3.1.2, and 3.1.3 briefly describe the ORACLES airborne sensors and their respective datasets, whereas section 5.1 presented the actual results of ACAOD from HSRL-2 and 4STAR, that are used in the above-cloud SSA inversion. We want to keep this distinction between the dataset description and their results.

l. 493: remove "noted"
Suggestion accepted.

l. 507. RMSD is not defined.
RMSD is now defined as the root-mean-square-difference.

The AERONET station Mongu is repeatedly mentioned and explained in the text. Remove all repetition.

We've removed the repletion as much as we could.

l. 607. Start a new paragraph here.
Suggestion accepted.

l. 630 lesser errors -> smaller errors.
Accepted.

l. 669 define PSD.
PSD is defined as the particle size distribution.

l. 730-735: Merge with the methods section (or simply remove this repetition).
We realize that the information provided here repeats what is already described earlier in the paper. So, we have accepted the reviewer's suggestion and removed line # 730-735 in the revision.

l. 774. one reason -> One reason of the bias
This should be line 744. The sentence corrected as "One reason for the bias, among several others…"

l. 776 CALOP -> CALIOP
Corrected.

References

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