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

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### **Response to the Anonymous Referee #1**

We thank and appreciate the anonymous reviewer for offering constructive and valuable comments & suggestions 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 significant revision stems from the comments made by referee # 1 regarding the cloud model properties. While 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 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  $\mu$ m with a wide distribution ranging from 6  $\mu$ m to 26  $\mu$ m, albeit 70%-80% of the data points are found to be within 6-18  $\mu$ m. 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  $\alpha$ ,  $\beta$ , and  $\gamma$  parameters of 6.0, 1.5, and 1.0, respectively. The CRE value, when calculated using these parameters, of this cloud model was 6  $\mu$ m, 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 in our study, we have revised the cloud model with an optimal CRE value of 12  $\mu$ m. This is achieved by changing the  $\alpha$  parameter from 6.0 to 15.0 and keeping the  $\beta$  and  $\gamma$  parameters unaltered.

The entire analysis presented in the initially 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 significant 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.

The following are the one-to-one responses to each comment offered by referee # 1.

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

**RC**: 1. Section 3.1: When the airborne measurements are introduced here, I suggest to add some descriptions about different flight modes of ER-2 and P3 since a few comparisons and different data processing methods are mentioned later. Actually, I don't fully understand the observation mode about how P3 aircraft profiled the atmosphere between the surface to 6 km. Did it only measure the atmosphere when flying vertically? What is the vertical resolution for the profiles?

**AR:** The ER-2 aircraft during the 2016 operation flew at about 20 km altitude and measured the vertically resolved aerosols and cloud properties for the atmospheric column below the aircraft level. On the other hand, P3 aircraft flew from the surface to about 5-6 km altitude, profiling the atmosphere vertically and horizontally within these altitude ranges. Different instruments onboard P3, including the 4STAR supphotometer, made measurements both in vertical and horizontal dimensions along the aircraft trajectories.

Pertaining to the columnar AOD measurements above the P3 altitude, 4STAR made measurements at a vertical resolution in the range of 1-3 meters during all three deployments with an average resolution of  $\sim$ 2 meters. There were also instances during the flights when P3 flew horizontally without changing altitude and measured spectral AODs.

**RC:** 2. Section 3.3.2: Should the "C1\*Alt" in the first two equations here be the "C1\*GPSAlt" and "C1\*CldTopAlt", respectively?

**AR:** That's true. '*Alt*' should be '*GPSAlt*' and '*CldTopAlt*' in the respective two equations. Thanks for pointing this out. The corrections are as follows.

 $AOD_{GPSAlt} = C0 + C1 * GPSAlt + C2 * GPSAlt^{2}$  $AOD_{CldTopAlt} = C0 + C1 * CldTopAlt + C2 * CldTopAlt^{2}$ 

**RC:** 3. Line 329-331: "The spectral dependence of the imaginary part of the refractive index in the visiblenear-IR wavelength range (470-860 nm) is described by the AERONET dataset" is mentioned here, but it is unclear that how to describe this spectral dependence. Is any linear or exponential assumption used?

**AR:** The AERONET Level 2.0 inversion algorithm retrieves both real and imaginary parts of the refractive index at four discrete wavelengths of 470 nm, 670 nm, 860 nm, and 1020 nm. Using the long-term inversion dataset of AERONET at an inland site, *Mongu*, for the biomass burning season (i.e., July, August, and September), we calculated the ratio of the imaginary part of the refractive index between 470 nm and 860 nm. We use this 470/860 nm ratio of the imaginary index to constrain the spectral dependence of absorption between 470-860 nm. Table 2 lists the imaginary index values at 470 nm and 860 nm, in which the values at 470 nm were selected in intervals and the same at 860 nm were calculated based on the derived ratio from the long-term AERONET climatology dataset.

**RC:** 5. Section 4.2: Some fixed microphysical parameters of cloud particles are mentioned here, including the effective radius, cloud layer height and cloud layer thickness. How will these parameters affect the aerosol-corrected COD retrieval? Given the effective radius comes from an old paper and the resources of cloud layer height and cloud layer thickness are not mentioned here, can they represent the climatology of clouds in the southeastern Atlantic Ocean? Maybe some references could be added.

**AR:** Please refer to our response at the beginning of this report on the significant modification applied to the revision. Briefly mentioning here, we've changed the cloud effective radius (CRE) from 6  $\mu$ m to 12  $\mu$ m of the adopted C1 cloud model in accordance with the MODIS CRE retrievals over the Southeastern Atlantic Ocean. We've also quantified and added the uncertainty estimates in the retrieved SSA and aerosol-corrected COD due to perturbation in CRE by  $\pm 6 \mu$ m. Please refer to Tables 3 to 6 in the revised paper.

Regarding the cloud layer height and thickness, we looked at the CALIOP/CALIOP Level -2 cloud product over the study region and derived the mean and median values of cloud top and cloud base altitudes for 2016 and 2017. The table below shows that the cloud top detected by CALIOP was between 1.2-1.4 km, whereas the cloud base was between 0.5-0.6 km above sea level. In the present study, we assumed the cloud top and bottom altitudes of 1.5 km and 1.0 km, respectively. While the assumed cloud top altitude is in close agreement with CALIOP, the cloud bottom is slightly overestimated by ~0.4 km. However, we don't expect any noticeable changes in the derived SSA and COD retrievals due to a slight departure of the cloud bottom altitude from the observations.

We have added this description in the revised paper in section 4.2.

Table 1 Cloud top and base altitudes (in km) over the Southeastern Atlantic Ocean (Lat: 33S to 0, Lon: 20W to 10E) detected by CALIOP/CALIPSO during biomass burning months of 2016 and 2017.

Months	Cloud Top (mean/median) [km]	Cloud Base (mean/median) [km]
August	1.36/1.31	0.58/0.53
September	1.35/1.31	0.64/0.62
October	1.26/1.22	0.49/0.47

**RC:** 6. Figure 6: Comparing Panel a and b, the SSA at 860 nm shows larger differences than that at 470 nm, even larger than other measurements in the rest of panels in this figure. Do you think what could be the possible reasons for this difference between HSRL-2 and 4STAR retrievals? Do you think what could be the possible reasons for this difference between HSRL-2 and 4STAR retrievals?

**AR:** Note that we have updated all figures with the revised results derived from the modified cloud model. In the revised Figure 6, the spread in the retrieved SSA at both 470 nm and 860 nm is noticed to be larger with the 4STAR synergy (panel b) than with the HSRL-2 synergy (panel a).

The proposed algorithm was applied identically to both HSRL-2 and 4STAR measurements, accompanied with synergized OMI and MODIS TOA reflectance observations. The only difference between the two sets of results is the kind of ACAOD datasets, i.e., lidar vs. sun photometer. The HSRL-2 directly measured the columnar AOD above the cloud and used as an input to the synergy algorithm. On the other hand, 4STAR onboard P3 measured columnar AOD above the aircraft level, which required an altitude-based adjustment to the observed cloud-top (see section 3.3.2) in order to establish an equivalence with satellite measurements. The adjustment procedure used flight-averaged altitude vs. AOD polynomial to achieve this job. Any departure of the actual altitude vs. AOD profile for individual 4STAR-satellite matchups from the averaged representation, therefore, leads to erroneous columnar AOD above clouds and retrieved SSA. Another reason for a relatively larger spread in the 4STAR synergy retrievals of SSA could be the different aircraft trajectories, and therefore airmass, sampled by both airborne instruments that could result in different ranges of aerosol absorption properties in the region.

# **RC:** 7. Section 7: The possible uncertainties in SSA retrievals from errors in aerosol properties are well analyzed in this section, but how will these properties affect aerosol-corrected COD?

**AR:** Thanks for pointing this out; we missed including the error matrix for COD due to changes in aerosol properties. In the revised manuscript, we have added additional Tables 5 and 6 containing the % error in the retrieved aerosol-corrected cloud optical depth at 388 nm and 470 nm, respectively, due to the presumed errors in input AOD, aerosol layer height, aerosol absorption Angstrom Exponent (AAE), particle size distribution, NO<sub>2</sub>, and cloud effective radius.

## Similarly, how is the sensitivity of aerosol-corrected COD with respect to the cloud properties in the cloud model?

To address the reviewer's concern, we also performed additional RT calculations of aerosols above cloud LUTs (both UV and VIS-NIR) with the C1 cloud model, but by perturbing the original cloud effective radius (CRE) of 12  $\mu$ m by  $\pm 6 \mu$ m, i.e., 6  $\mu$ m and 18  $\mu$ m. The new set of aerosol-cloud LUTs was used to re-retrieve SSA and aerosol-corrected CODs for comparison with the original set of retrievals.

The changes in the retrieved SSA and COD are appended in Tables 3 through 6 in the revision. To summarize the findings, a decrease (an increase) in CRE by 6  $\mu$ m produces a -8.67% (+2.39%) error in the retrieved COD at 388 nm. The errors in the retrieved COD at 860 nm, however, were higher and -13.29%

(6.17%). Larger (relatively smaller) errors in COD at longer (shorter) wavelength (388 nm and 860 nm in this case) imply greater (relatively weaker) sensitivity of the TOA reflectance and retrieved COD to the change in liquid water droplet distribution.

We've added a new sub-section 7.7 describing the sensitivity of the retrieved COD to various assumptions made in the inversion algorithm.

The new results are described in section 7 in the revision.