

Subseasonal and spatial variability of biomass burning aerosol radiative properties observed over the Southeast Atlantic during ORACLES 2016-2018

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Abstract. During 2016-2018, NASA conducted the ORACLES (ObseRvations of Aerosols above CLouds and their intErActionS) airborne field campaigns to study aerosol-cloud-radiation interactions with the stratocumulus cloud deck over the Southeast Atlantic. ORACLES employed 4STAR (Spectrometer for Sky-Scanning, Sun-Tracking Atmospheric Research) to measure direct solar irradiances and diffuse sky radiances from which free-tropospheric Biomass Burning Aerosols (BBA) radiative properties are derived. Changes in Single Scattering Albedo (SSA) indicate increased scattering as the biomass burning season progresses, which we attribute to an aerosol brightening from compositional changes, rather than a change in aerosol type, with an apparent lack of Brown Carbon throughout the season. A collection of 31 AERONET (AErosol RObotic NETwork) sun/sky photometer stations have operated in Southern Africa for over thirty years (1995-2025), creating a complete aerosol climatology for the first time, which can be compared with SSA and Aerosol Optical Depth (AOD) from ORACLES observations. The spatial distributions of in situ SSA are also investigated by latitude, longitude, and altitude. The latitudinal analysis indicates that the September decrease in SSA noted by 4STAR is affected by the southerly sampling location of ORACLES 2016 compared to the other two campaigns. Westward gradual increases and sharper decreases in SSA are attributed to late-transport aging processes identified by previous studies. These processes start further eastward in October, in conjunction with the southeastward shift in source fires. Collectively, ORACLES 4STAR retrievals and in situ measurements have identified subseasonal and spatial trends in SSA over the Southeast Atlantic that complement the Southern African AERONET climatology.

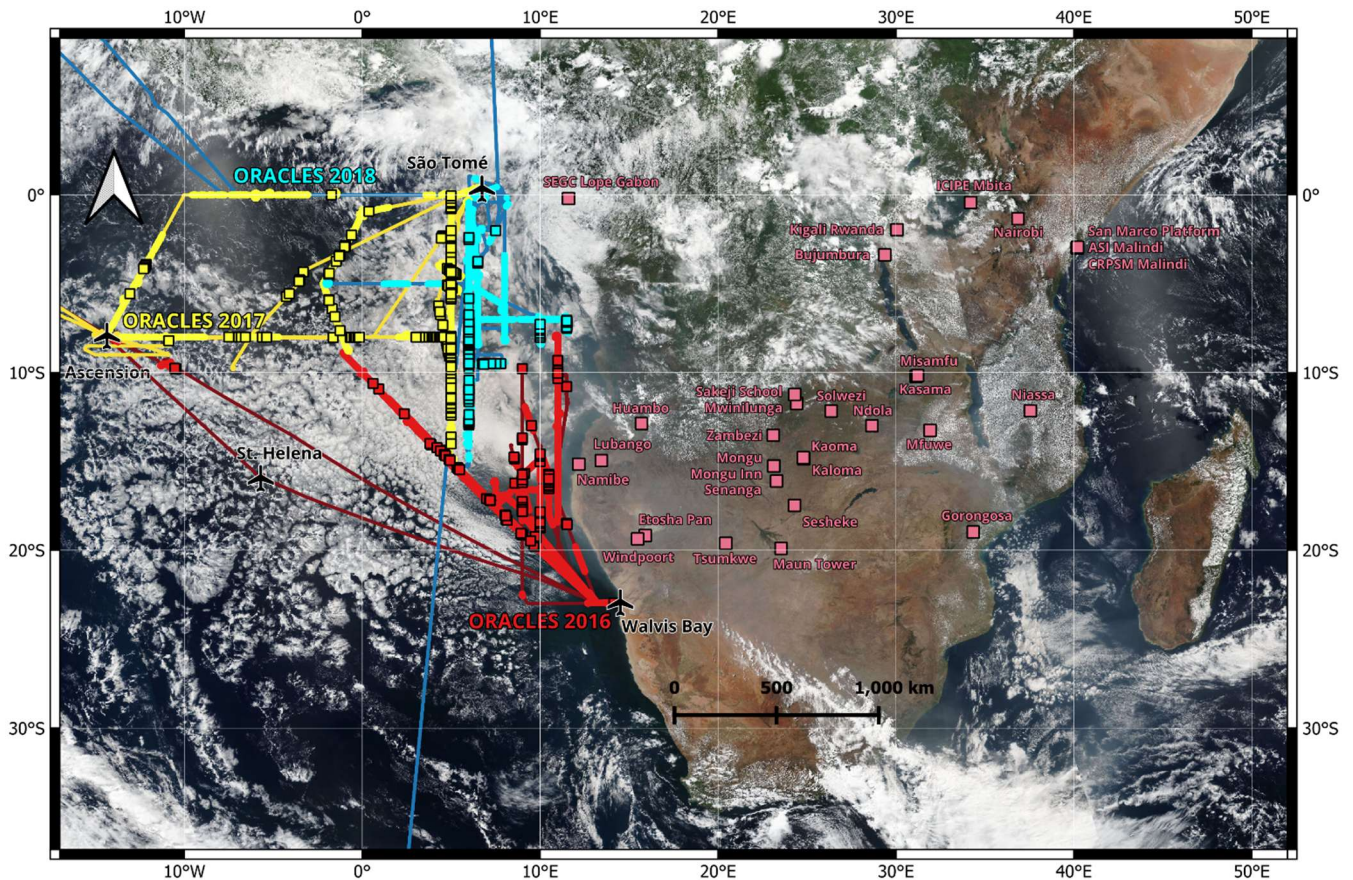
29 1 Introduction

30 The Southeast Atlantic Ocean (SEA) is a region with a high degree of uncertainty regarding aerosol-radiation and
31 aerosol-cloud interactions (Meyer et al., 2013; de Graaf et al., 2014; Mallet et al., 2020; Brown et al., 2021; Che et al., 2021).
32 A major reason for this is the difficulty involved with observing aerosols in the region, especially due to the presence of a
33 semi-permanent subtropical stratocumulus cloud deck (Sakaeda et al., 2011; Waquet et al., 2013). Satellite-based aerosol
34 retrievals, such as those employed by MODIS (MODerate resolution Imaging Spectroradiometer) onboard the Aqua and
35 Terra satellites, are limited by their simplified reflectance-based atmospheric retrieval algorithms when applied to complex
36 atmospheric conditions. In the case of the SEA, aerosol smoke plumes overlie the stratocumulus cloud deck, making it
37 difficult for satellites to discern the above-cloud aerosol properties (Jethva et al., 2024).

38 The remoteness of the SEA also makes it difficult to employ ground-based observations within the region.
39 AERONET (AErosol RObotic NETwork), a global confederated network of ground-based Cimel sun/sky photometers for
40 the observation of columnar aerosol properties (Holben et al., 1998), is limited to just three stations in the SEA - Ascension
41 Island, St. Helena, and São Tomé. As such, three widely spaced stations yield insufficient resolution to capture most smoke
42 events in the region. As part of the international concerted efforts to study the aerosol-cloud system over the SEA, many
43 AERONET stations within the aerosol source region of Southern Africa were established (Redemann et al., 2021), but the
44 network still has sizeable gaps. The first AERONET station in Southern Africa was established in Mongu, Zambia (15.25 °S,
45 23.15 °E) in 1995, recently allowing for a full thirty-year aerosol climatology of that region for the first time. Thus, airborne
46 campaigns provide a critical link for studying aerosols over the SEA by synergizing with other insights from ground-based
47 measurements and satellite-based observing systems (Zuidema et al., 2016; Zuidema et al., 2018; Formenti et al., 2019;
48 Haywood et al., 2021; Redemann et al., 2021).

49 ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) was a NASA Earth Venture
50 Suborbital field campaign to study Biomass Burning Aerosols (BBA) over the SEA from 2016 to 2018 (Redemann et al.,
51 2021), using a suite of instruments aboard a P-3 Orion aircraft (2016-2018) and a high-flying ER-2 aircraft (2016 only). One
52 of the instruments deployed on the P-3 aircraft was 4STAR (Spectrometer for Sky-Scanning, Sun-Tracking Atmospheric
53 Research), a sun/sky spectrophotometer for retrieving columnar aerosol radiative properties (Pistone et al., 2019; LeBlanc et
54 al., 2020), serving as an airborne complement to ground-based AERONET retrievals. The campaigns were approximately a
55 month long each and occurred during August 2017, September 2016, and October 2018, collectively covering the peak of
56 Southern Africa's BBA emission season (Queface et al., 2011), which contributes approximately 35 % of global BBA
57 emissions (van der Werf et al., 2010; Granier et al., 2011). In 2016, ORACLES was based out of Walvis Bay, Namibia,
58 while in 2017 and 2018, it was based out of São Tomé (Fig. 1). BBA smoke plumes are transported from mainland Africa to
59 the SEA by the Southern African Easterly Jet (Adebisi and Zuidema, 2016; Holanda et al., 2020), with a maximum jet
60 strength at 3 km in August and 4 km in September and October (Ryoo et al., 2021). Once over the SEA, the BBA overlies
61 and interacts with a semi-permanent subtropical stratocumulus cloud deck (Sakaeda et al., 2011; Wilcox, 2012; Waquet et

62 al., 2013). There is remarkably little interannual variability in Southern African fires due to the consistency of agricultural
63 burning practices in the region (van der Werf et al., 2010; Redemann et al., 2021). Using the 18-year (2001–2018) European
64 Space Agency Fire Climate Change Initiative, version 5.1 (FireCCI51) dataset derived from MODIS aboard the NASA Terra
65 satellite, Chuvieco et al. (2021) found that Southern Africa had the smallest interannual variability in burned area in the
66 world, with a coefficient of variation < 0.5 across the region. Ryoo et al. (2021) documented the interannual variability in
67 meteorological conditions over the Southeast Atlantic, finding that wind velocity, relative humidity, and precipitation were
68 near climatological averages for August 2017, September 2016, and October 2018. They also found that despite sea surface
69 temperatures during the ORACLES campaign months being $0.2 - 0.4$ K above climatological averages, they did not have a
70 systematic effect on low-level cloud fraction.



71
72 **Figure 1: The ORACLES 2016–2018 P-3 flight paths over the Southeast Atlantic and the 31 AERONET stations in Southern**
73 **Africa utilized in this study. ORACLES flight paths are dark lines colored by campaign, with brighter circles for valid in situ**
74 **measurements and brighter squares for valid 4STAR sky-scans. Pink squares represent AERONET stations. The ORACLES**
75 **headquarters in Walvis Bay, Namibia (2016) and São Tomé (2017–2018), as well as refuelling stations at St. Helena and Ascension**
76 **Island are highlighted using airplane symbols. ORACLES 2018 data was transposed 1° eastward as to distinguish it from**
77 **ORACLES 2017 data, making the north-south transit leg at 5° E visible for both campaigns. Satellite imagery is true color**
78 **corrected reflectance from the VIIRS (Visible Infrared Imaging Radiometer Suite) aboard the NASA/NOAA Suomi satellite for 12**
79 **September 2016, courtesy of NASA Worldview.**

80 BBA is the largest global source of carbonaceous aerosols (Bowman et al., 2009) and can be composed of strongly
81 absorbing elemental aerosols called Black Carbon (BC), less-absorbing organic aerosols called Brown Carbon (BrC), and
82 non-absorbing Organic Carbon. The full extent of BrC's contribution to both global and regional BBA absorption is the
83 subject of ongoing research (Andreae and Gelencsér, 2006; Zhang et al., 2020). Previous studies have concluded that little
84 BrC is present over the SEA (Denjean et al., 2020; Taylor et al., 2020; Wu et al., 2020; Dobracki et al., 2023; Dobracki et al.,
85 2025), with Taylor et al. (2020) estimating that BrC only accounted for 9 to 11 % of absorption at 405 nm. Using TEM-EDX
86 (Transmission Electron Microscopy - Energy Dispersive X-ray spectroscopy) and an assumed BC refractive index, Zhang et
87 al. (2022) determined that BrC could contribute 8 – 22 % of total aerosol absorption at 470 nm. However, their BrC estimate
88 is much lower using the AAE attribution method, at only 2 – 6 % of total aerosol absorption at 470 nm, indicating that
89 spectroscopy could reveal a greater BrC presence over the Southeast Atlantic than suggested by optical methods. However,
90 many studies lack optical measurements within the UV spectrum, where BrC absorption is greatest, allowing questions
91 regarding the potential presence of BrC to persist.

92 Absorbing aerosols cause a decrease in environmental albedo that is strongest over brighter surfaces, while non-
93 absorbing aerosols cause an increase in environmental albedo over darker surfaces (Bellouin et al., 2020). Absorbing
94 aerosols generally cause negative radiative forcing (cooling) over the SEA in cloudless conditions but can cause positive
95 radiative forcing (warming) when above a stratocumulus cloud deck (Keil and Haywood, 2003; Chand et al., 2009). BBA
96 interactions with the stratocumulus cloud deck over the SEA are considered a major source of uncertainty for climate
97 modelling of the region (Stier et al., 2013; Zuidema et al., 2016; Haywood et al., 2021; Doherty et al., 2023), in that models
98 show a large spread in predictions and exhibit significant discrepancies relative to observations.

99 The goal of this study is to determine the subseasonal changes in BBA radiative properties that occurred in
100 ORACLES 2016-2018, within the Southern African BBA emission season of August to October. We examine Single
101 Scattering Albedo (SSA) and Aerosol Optical Depth (AOD), the two aerosol properties most pertinent to modellers, but also
102 discuss Aerosol Absorption Optical Depth (AAOD), Extinction Ångström Exponent (EAE), Absorption Ångström Exponent
103 (AAE), and complex refractive indices as well, for a comprehensive view of aerosol radiative changes. SSA from airborne
104 4STAR sky-scans and in situ measurements are compared to assess agreement within the ORACLES 2016-2018 campaigns
105 regarding any apparent subseasonal trends. We also conduct spatial analyses (including latitudinal, longitudinal, and
106 altitudinal) on the in situ SSA data, as to further isolate subseasonal variability from other effects due to chemical and
107 physical processing. The spatial analyses will help to explain apparent discrepancies between datasets, as each ORACLES
108 campaign differs in the latitudes, longitudes, and altitudes sampled. SSA and AOD from ORACLES are also compared
109 against AERONET, allowing for comparison of the BBA radiative properties from the SEA study region and the Southern
110 African source region, as well as a comparison against a full aerosol climatology for the first time.

111 2 Data and Methods

112 2.1 4STAR

113 4STAR is a sun/sky spectrophotometer measuring direct beam solar irradiances and diffuse sky radiances (Dunagan
114 et al., 2013), designed as an airborne complement to ground-based AERONET Cimel instruments. During ORACLES,
115 4STAR was mounted to the top of the NASA P-3 Orion aircraft, where it observed above-cloud, below-plume columnar
116 aerosol radiative properties (Pistone et al., 2019; LeBlanc et al., 2020). 4STAR also features hyperspectral resolution
117 measurements that permit the retrieval of some gas species (Segal-Rozenhaimer et al., 2014). Direct solar irradiances and
118 diffuse sky radiances measured by 4STAR, along with flight-level albedo calculated from SSFR (Solar Spectral Flux
119 Radiometer) measurements of nadir upwelling and zenith downwelling spectral irradiances (Coddington et al., 2008;
120 Schmidt and Pilewskie, 2012; Cochrane et al., 2019; Cochrane et al., 2021; Schmidt et al., 2021; Cochrane et al., 2022;), are
121 used as inputs in an aerosol inversion code adapted from AERONET version 2.0 (Holben et al., 2006; Dubovik and King,
122 2000; Holben et al., 1998) for airborne use. This code allows us to retrieve aerosol radiative properties from 4STAR and
123 SSFR measurements, including SSA, AOD, AAOD, EAE, AAE, RRI, and IRI. SSA, the ratio of aerosol scattering to total
124 aerosol extinction, is one of the most important variables for determining an aerosol's radiative impacts (Takemura et al.,
125 2002; Bergstrom et al., 2007; Moosmüller et al., 2012). The SSA of BBA emitted from Southern Africa can quickly evolve
126 (Abel et al., 2003), requiring highly accurate observations, preferably over the full aerosol life cycle. AOD measures the
127 columnar extinction of light by aerosols, while AAOD measures the columnar absorption of light by aerosols. EAE is the
128 spectral dependence of AOD, with AAE being the spectral dependence of AAOD. The complex refractive indices are one of
129 the primary outputs of the 4STAR retrievals, which are used to iteratively reduce retrieval errors, with the RRI representing
130 aerosol scattering and IRI representing aerosol absorption. 4STAR sky radiance measurements were calibrated in the
131 laboratory using an NIST (National Institute of Standards and Technology) referenceable 12-lamp 36 in. (91.44 cm)
132 integrating sphere (Brown et al., 2005). Whereas 4STAR direct beam measurements were calibrated via refined Langley
133 regressions (Schmid and Wehrli, 1995) conducted at Mauna Loa Observatory, before and after deployment (LeBlanc et al.,
134 2020; Mitchell et al., 2025).

135 We utilized 4STAR sky-scans to determine columnar aerosol properties for ORACLES 2016-2018. Our Quality
136 Control (QC) criteria for archival in the NASA ASDC (Atmospheric Science Data Center) repository were:

- 137 (1) AOD (400 nm) > 0.2,
- 138 (2) altitude difference < 50 m,
- 139 (3) mean sky radiance error ($|\text{measured} - \text{fit}|$) < 10 %,
- 140 (4) minimum scattering angle < 6 °,
- 141 (5) maximum scattering angle > 50 °,
- 142 (6) mean scattering angle difference < 3 °,
- 143 (7) maximum scattering angle difference < 10 °,

(8) covariance matrix sky error < 10 %,

(9) roll standard deviation < 3 °, and

(10) passes the retrieval boundary test.

Criteria (6) and (7) are only considered over the critical scattering angle range of 3.5 - 30 °. For this study, we additionally required that all sky-scans were taken from flight altitudes below 3 km, as to best ensure the full column of the smoke plume was observed. These sky-scans retrieve at four wavelengths (500, 675, 870, 995 nm), differing slightly from the five-wavelength set (400, 500, 675, 870, 995 nm) used by Pistone et al. (2019). We excluded 400 nm due to the presence of an instrument artifact (Mitchell et al., 2025) that affected wavelengths near 420 nm in 2017 and 2018. For uniformity, this was applied to ORACLES 2016 as well. These sky-scans are the T0_2016_4wl, R0_2017_4wl, and R0_2018_4wl datasets meeting Selection #2 criteria, as described in Table 2 of Mitchell et al., 2025. The 4STAR sky-scans are summarized in Table 1, along with comparable in situ measurements and ground-based AERONET observations, which are also detailed in the following subsections.

Table 1: The instruments and variables used in this study, along with their reporting wavelengths and valid observation counts. The PSAP (Particle Soot Absorption Photometer) & Neph (Nephelometer) SSA observation counts are following the interpolation of scattering coefficients from the Neph wavelengths (450, 550, 700 nm) to the PSAP wavelengths (470, 530, 660 nm). The given PSAP & Neph observation count is for 530 nm, with slightly more measurements at 470 nm, and slightly fewer at 660 nm. The AERONET observation counts are from the 31 Southern African stations displayed in Fig. 1, from 01 July to 31 October, for 1995-2025.

Instrument	Platform	Pertinent Variables	Wavelengths (nm)	Observation Count
4STAR (& SSFR)	ORACLES P-3	SSA, AOD, AAOD, EAE, AAE, RRI, and IRI from sky-scans (using flight-level albedo from SSFR)	500, 675, 870, 995	77 (2016) 141 (2017) 92 (2018)
PSAP & Neph	ORACLES P-3	SSA from in situ PSAP absorption and Neph scattering coefficients	PSAP: 470, 530, 660 Neph: 450, 550, 700	1.1×10^5 (2016) 6.4×10^4 (2017) 7.6×10^4 (2018)
AERONET	Ground	SSA from sky-scans AOD from direct beam measurements	440, 675, 870, 1020	5.0×10^4 (SSA) 5.0×10^5 (AOD)

2.2 In Situ Measurements

During ORACLES, in situ instruments aboard the P-3 Orion aircraft were operated by the HIGEAR (Hawaii Group for Environmental Aerosol Research) team. This includes two TSI 3563 nephelometers (Neph), which determine in situ aerosol scattering of light. Scattering coefficients are measured at three wavelengths (450, 550, 700 nm) using Anderson and Ogren (1998) corrections that account for Scattering Ångström Exponent (SAE) and depend upon both wavelength and the presence/lack of a submicron impactor. Two Radiance Research PSAPs (Particle Soot Absorption Photometers), also operated by HIGEAR aboard the P-3 aircraft, were used to determine in situ aerosol light absorption. Absorption coefficients are measured at three wavelengths (470, 530, 660 nm) using Virkkula (2010) wavelength-averaged corrections from the fittings of transmission correction functions, black aerosol experiments, and ammonium sulfate experiments.

A newly processed in situ SSA dataset for ORACLES 2016-2018 was also used in this study. SSA was calculated from 1 Hz Neph scattering coefficients (σ_{scat}) and PSAP absorption coefficients (σ_{abs}) that had been subjected to a 10 s

rolling average. A scattering coefficient screening of $> 10 \text{ Mm}^{-1}$ was applied per wavelength to reduce statistical noise, such that SSA_{470} requires $\sigma_{\text{scat}_470} > 10 \text{ Mm}^{-1}$, SSA_{530} requires $\sigma_{\text{scat}_530} > 10 \text{ Mm}^{-1}$, and SSA_{660} requires $\sigma_{\text{scat}_660} > 10 \text{ Mm}^{-1}$. Additional screenings were also applied to target the free-tropospheric BBA smoke plume (carbon monoxide concentrations $> 130 \text{ ppbv}$ and altitude $> 1.5 \text{ km}$) allowing for comparison with 4STAR. BBA is the dominant aerosol present in the free troposphere over the SEA, but it may be mixed with or dominated by sea salt aerosols within the boundary layer (Dang et al., 2022), warranting the latter's exclusion. Due to the 10 s rolling average, when using the in situ data for statistical analyses, we assume that the number of independent samples is one tenth of the observation count. Given that the typical ORACLES sampling speed of the NASA P-3 Orion aircraft was $\sim 382 \text{ km per hour}$, this results in a spatial resolution of about $1.1 \text{ km per independent sample}$.

The ORACLES campaigns span 09 August to 02 September 2017, 27 August to 27 September 2016, and 24 September to 25 October 2018. For simplicity, we will often summarize the ORACLES 4STAR and in situ data per campaign, which we will refer to as “months”, as most research flights occurred in August 2017, September 2016, and October 2018, respectively. For valid 4STAR retrievals, only one research flight was outside of the primary month per campaign. It is a similar case for valid in situ measurements, except that ORACLES 2018 had two research flights outside of the primary month.

2.3 AERONET

AERONET cloud-screened (level 1.5) AOD from direct beam measurements and SSA from almucantar and hybrid sky-scans (Giles et al., 2019; Sinyuk et al., 2020) were obtained for 31 Southern African stations (Fig. 1) for 1995-2025 to represent an aerosol climatology of ground-based measurements of similar data quality to 4STAR. The primary purpose of analyzing the climatology is to assess the representativeness of the three-year ORACLES period. This dataset was then limited to 2016-2018, so that just the concurrent ORACLES years could be compared as well, which contains approximately 31 % of the data within the full climatology.

When selecting AERONET stations, we limited the latitudinal range to $0^\circ - 20^\circ\text{S}$, targeting only the near-source AERONET stations upwind of the SEA, while aiming to avoid any anticyclonic recirculations that transport BBA back towards the southerly latitudes of mainland Africa (Adebiyi and Zuidema, 2016; Redemann et al., 2021). These latitudinal restrictions were not applied to ORACLES 2016 in situ data so that its full latitudinal dependence could be observed, while only one valid 4STAR sky-scan occurred south of 20°S .

3 Results

3.1 SSA Subseasonal Analysis

4STAR radiative properties, including SSA, AOD, AAOD, EAE, AAE, and complex refractive indices, are organized by ORACLES campaign month (August 2017, September 2016, October 2018) and box-plotted to illustrate their

205 subseasonal variabilities. All four wavelengths are displayed within each campaign, so that their spectral dependence can
206 also be observed. For SSA, in situ measurements at all three PSAP wavelengths are also included.

207 Both 4STAR retrievals and in situ measurements exhibit an increase in median SSA over the course of the BBA
208 emission season (Fig. 2), with the increase from August to October ranging from +0.02 at longer wavelengths to +0.04 at
209 shorter wavelengths. 4STAR displays a decrease in SSA medians from August to September not found in the in situ data,
210 which we address later using spatial analyses. The 4STAR SSA variability of 0.79-0.95 at 500 nm for ORACLES 2017
211 aligns with that found by Cochrane et al. (2021) using an SSFR absorption-derived method. Applying a two-sample
212 Wilcoxon rank sum test (which determines if the samples are from continuous distributions with equal medians) to the in situ
213 data (reduced to the independent sample size) yields p-values < 0.001 for both monthly transitions (August-September and
214 September-October) at all three PSAP wavelengths, indicating a strongly significant increase through the season. Applying
215 the same test to the 4STAR sky-scans for the transition from August to September yields p-values < 0.05 at 500 and 675 nm,
216 with p-values of 0.17 and 0.07 at 870 and 995 nm, while September to October yields p-values < 0.001 at all four 4STAR
217 wavelengths. This indicates that the August to September decrease only recorded by 4STAR is not statistically significant at
218 the longer wavelengths, whereas the September to October increase is strongly significant like that found in the in situ data.

219 This shows an increase in aerosol scattering, relative to total extinction, suggesting aerosol brightening as the
220 season progresses. An SSA increase of this magnitude has a complex and significant impact on variables related to aerosol-
221 radiation interactions, including the Direct Aerosol Radiative Effect (DARE) and heating rates, so it needs to be properly
222 accounted for in aerosol models. An increase in mid-visible median SSA from 0.84 in August to 0.88 in October can cause
223 critical albedo (the scene albedo whereby an overlying aerosol layer switches from warming to cooling) to increase from
224 0.23 to 0.27 (Cochrane et al., 2021). The critical albedo's sensitivity to SSA is particularly important for clear and partially
225 cloudy conditions with flight-level albedos less than 0.25, with heightened SSA in October decreasing the chances for
226 aerosol warming. This implies that heightened SSA in October is likely to weaken DARE and reduce heating rate efficiency
227 aloft (Cochrane et al., 2022), which in turn, would decrease atmospheric stability. This SSA increase has been noted by
228 previous studies and is likely linked to the southeastward shift in fires (Eck et al., 2013; Redemann et al., 2021; Choi et al.,
229 2024; Tatro and Zuidema, 2025), although the exact physical cause has been unclear.

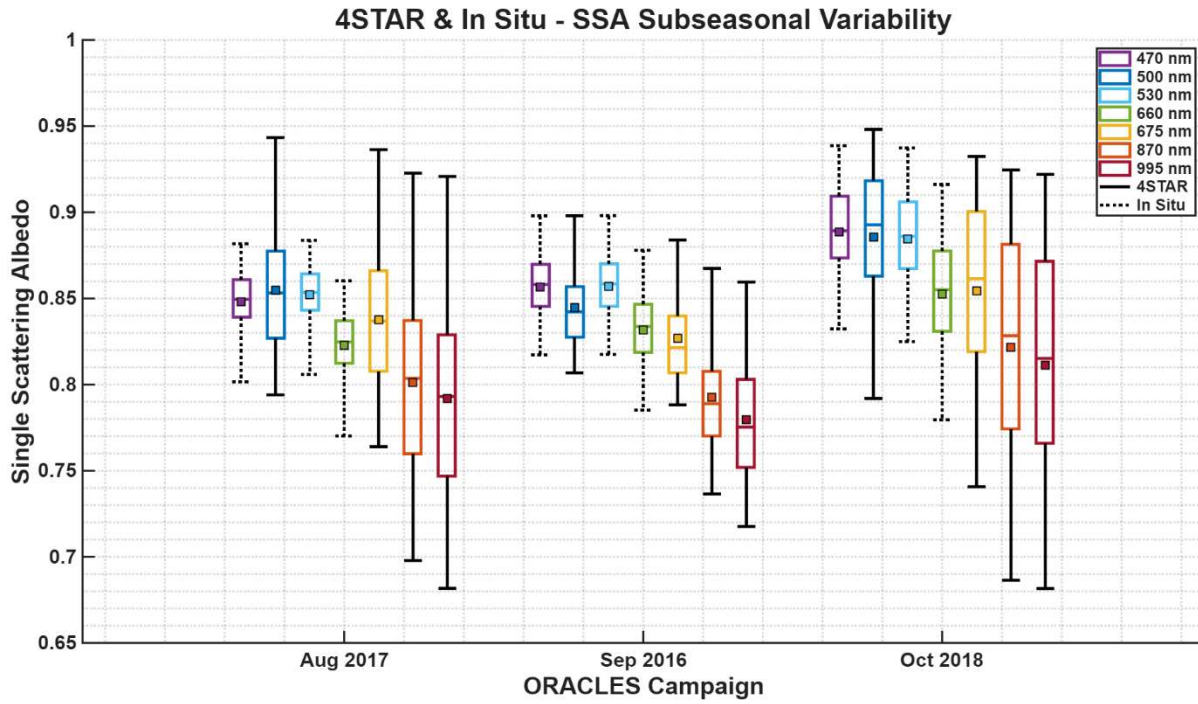


Figure 2: Subseasonal variability of 4STAR (solid) and in situ (dashed) SSA from ORACLES 2016-2018. Boxes show the interquartile range, while whiskers extend to the 5th and 95th percentiles. Central lines are medians and squares are means.

3.2 AOD and AAOD Subseasonal Analysis

At 500 nm, median AOD (and standard deviations) increases from 0.36 in August to a peak of 0.40 in September, then decreases to 0.31 in October, with similar relative changes at longer wavelengths (Fig. 3). Applying a two-sample Wilcoxon rank sum test to AOD, the transition from August to September yields p-values < 0.05 , while September to October yields p-values < 0.001 (both considering all four wavelengths). This indicates that the August to September AOD increase is moderately significant, whereas the September to October decrease is strongly significant. Although burned area in Southern Africa peaks in August, previous studies have found that persistent aerosol accumulation in the atmosphere delays the peak in aerosol loading until September (Eck et al., 2013; Adebisi et al., 2015; Redemann et al., 2021; Tatro and Zuidema, 2025). 4STAR AOD being greatest in September reflects these previous findings, but the transition from August is less dramatic, likely due to the lack of sampling in early August, when AOD would be lower.

AAOD parallels the trend in AOD, with median values at 500 nm of 0.055 in August, 0.069 in September, and 0.033 in October, which is again reflected at longer wavelengths as well. Applying the two-sample Wilcoxon rank sum test to AAOD, the transition from August to September yields p-values < 0.005 , while September to October yields p-values < 0.001 (again considering all four wavelengths). This indicates that the initial AAOD increase is strongly significant, while the subsequent AAOD decrease has even greater significance. The similarity to the AOD trend is somewhat unsurprising, as AAOD is also an extensive property and thus dependent upon aerosol loading, but the correlated behavior suggests that there

is not a significant change in aerosol type, which will be further verified using EAE and AAE in the next section. The decrease in AAOD from September to October is concurrent with the SSA increase from Fig. 2, indicating an absorption decrease coincident with the aerosol brightening.

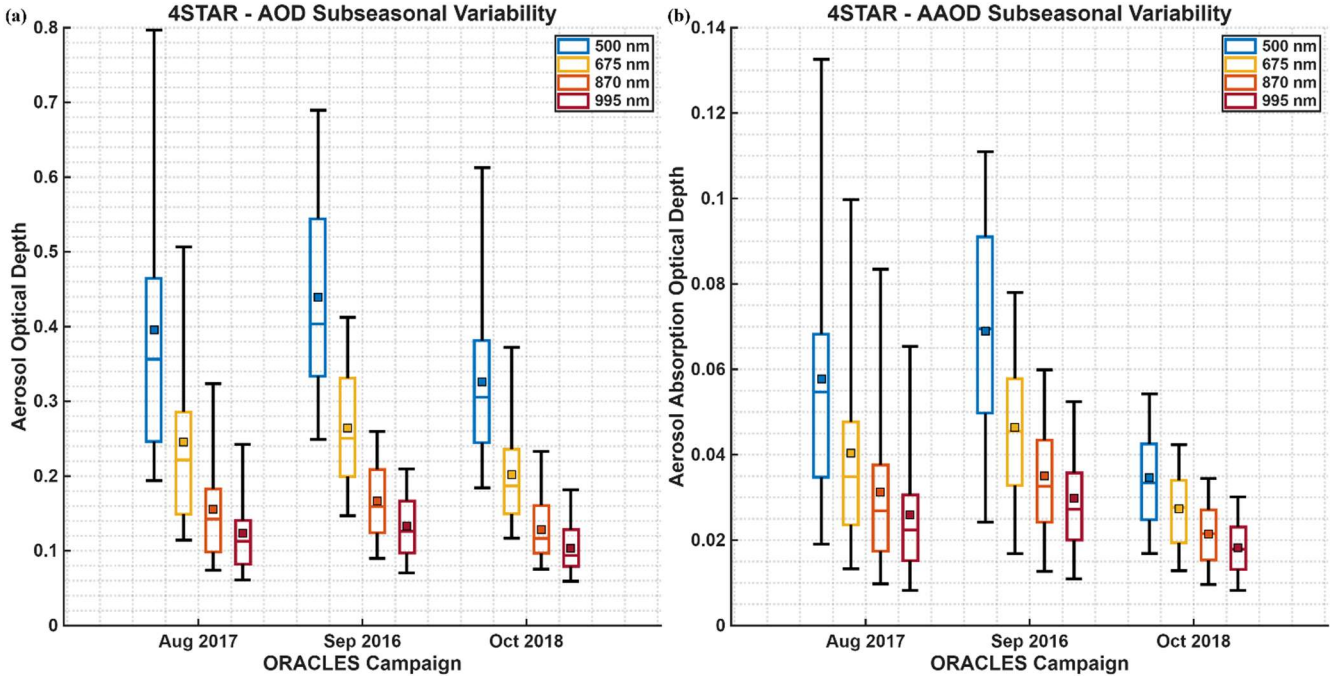
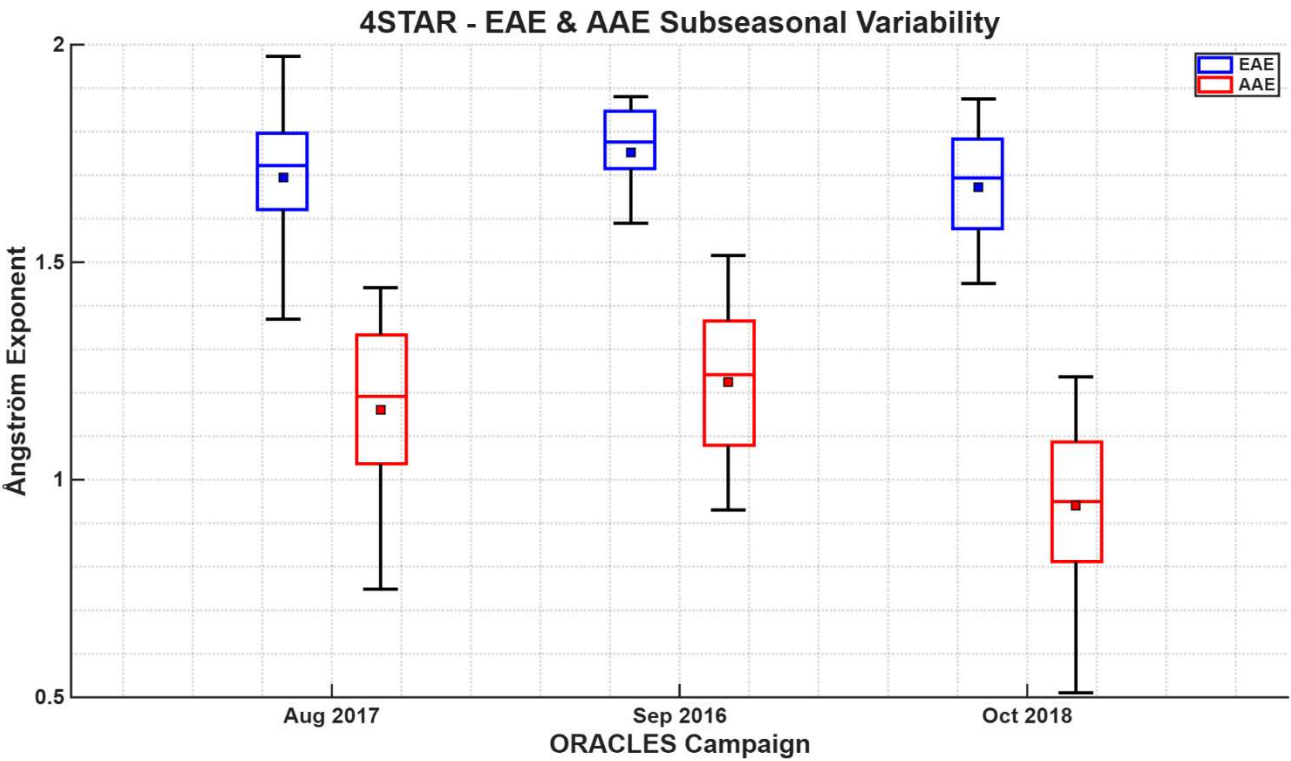


Figure 3: Subseasonal variability of 4STAR (a) AOD and (b) AAOD from ORACLES 2016-2018. Boxes show the interquartile range, while whiskers extend to the 5th and 95th percentiles. Central lines are medians and squares are means.

3.3 EAE and AAE Subseasonal Analysis

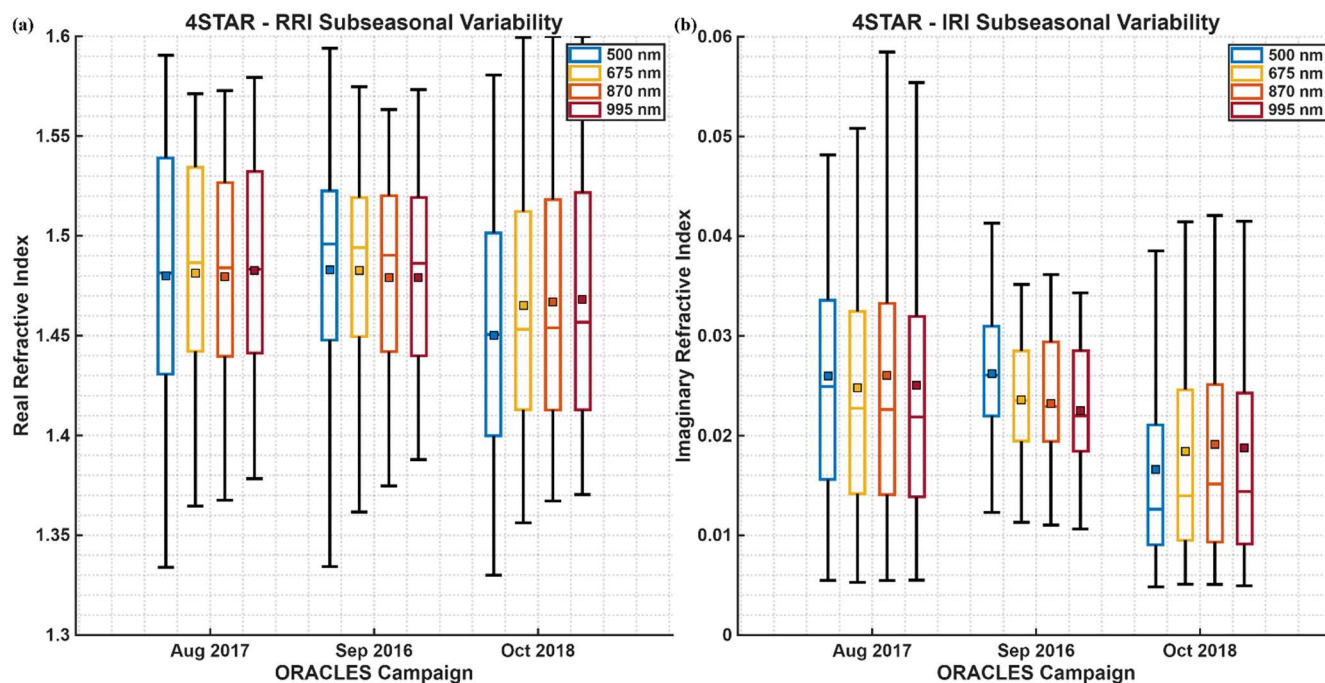
EAE and AAE were calculated by performing log-log spectral linear regressions of AOD and AAOD, respectively, using all four wavelengths (500, 675, 870, 995 nm) as input. The EAE campaign medians are centered near 1.73 (Fig. 4), with about 90 % of the total distribution falling within the 1.5 - 2.0 range, indicating fine aerosols (Schuster et al., 2006). There is greater variability within the AAE distributions, which is expected given its sensitivity to the much smaller absorption component. The AAE interquartile ranges are about 1.05 - 1.35 for August and September, which drops to 0.8 - 1.1 in October, suggesting the dominance of BC throughout the season. The presence of BrC cannot be fully discounted, as this study was unable to directly examine the UV wavelengths that are best absorbed by BrC (Alexander et al., 2008), but the lack of BrC contributions does align with previous findings (Denjean et al., 2020; Taylor et al., 2020; Wu et al., 2020; Dobracki et al., 2023; Dobracki et al., 2025). The lack of BrC contributions from our AAE analysis also aligns with the findings of Zhang et al. (2022), although their results indicate that electron microscopy would yield greater BrC contributions than optical methods. However, if the aerosol brightening in October was caused by increased BrC contributions, we would expect the AAE to increase, rather than decrease (Russell et al., 2010). Since there does not appear

268 to be significant changes in aerosol types during the BBA emission season, such as the processes described by Bond et al.
 269 (2013), we conclude that the aerosol brightening is instead due to changes in aerosol composition (Eck et al., 2013). This is
 270 supported by BC and OC columnar mass densities from MERRA-2 (Modern-Era Retrospective analysis for Research and
 271 Applications, version 2) reanalysis (Fig. S1), where the BC:OC ratio remains near 1:10 throughout the ORACLES campaign
 272 months, indicating little change in BrC concentration. August has the greatest BC and OC densities, although the maximums
 273 are close together and located over land. By September, higher BC and OC densities are more widespread across the
 274 Southeast Atlantic, followed by decreases in October, aligning with the September peak in AOD and AAOD noted by
 275 4STAR. BBA composition is dependent upon both fuel type and burning conditions, with the less-efficient smoldering fire
 276 phase generating more organic carbon and less BC than during the flaming phase (Levin et al., 2010; Collier et al., 2016; Jen
 277 et al., 2019; Dobracki et al., 2025). Back-trajectories performed by Dobracki et al. (2023) source less-scattering September
 278 BBA from grass fuels in the Miombo woodlands of Angola. The source fires then shift southeastward in October to
 279 Botswana, Zambia, Zimbabwe, and Mozambique, in alignment with the higher SSA values observed in that region (Eck et
 280 al., 2013; Choi et al., 2024; Tatro and Zuidema, 2025). A similar southeastward shift over the course of the BBA emission
 281 season is seen in the BC and OC columnar mass densities (Fig. S1), as well as in MODIS-detected vegetation fires (Fig. S2),
 282 and in HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) back-trajectories (Fig. S3).



283
 284 **Figure 4: Subseasonal variability of 4STAR EAE (blue) and AAE (red) from ORACLES 2016-2018. Boxes show the interquartile**
 285 **range, while whiskers extend to the 5th and 95th percentiles. Central lines are medians and squares are means.**

287 We also examined the subseasonal variability of the complex refractive indices (Fig. 5), including both RRI and
 288 IRI. At 500 nm, median RRI increases from about 1.48 in August to 1.50 in September, then falling to 1.45 in October, with
 289 similar relative changes at longer wavelengths. Applying a two-sample Wilcoxon rank sum test to RRI, the transition from
 290 August to September is not statistically significant at any wavelength, while September to October yields p-values < 0.05 at
 291 500 and 675 nm, with p-values of 0.10 and 0.14 at 870 and 995 nm. The IRI trend appears similar, with the IRI median at
 292 500 nm increasing from about 0.025 in August to 0.026 in September, and falling to 0.013 in October, which is again
 293 reflected at the longer wavelengths. Applying the same test to IRI, the transition from August to September is once again not
 294 statistically significant, but the transition from September to October yields p-values < 0.001 (both considering all four
 295 wavelengths). The lack of an increase in RRI coupled with a strong decrease in IRI suggests that the aerosol brightening in
 296 October is dependent upon a decrease in aerosol absorptivity, rather than an actual increase in its scattering.



297
 298 **Figure 5: Subseasonal variability of 4STAR (a) RRI and (b) IRI from ORACLES 2016-2018. Boxes show the interquartile range,**
 299 **while whiskers extend to the 5th and 95th percentiles. Central lines are medians and squares are means.**

300 4 Discussion

301 4.1 ORACLES and AERONET

302 To compare SSA and AOD from ORACLES and AERONET, we updated and expanded upon Fig. 3 from
 303 Redemann et al. (2021). AERONET observations are discretized into cells of 0.005 SSA (or 0.05 AOD) per day, then

304 colored by median latitude to emphasize which stations are most contributing to the dataset, with opacity weighted by the
305 number of observations. 10-day SSA and AOD medians are calculated from the AERONET observations, centered on Julian
306 dates ending in five. The same method was also applied to determine 10-day medians from ORACLES in situ SSA. SSA and
307 AOD from 4STAR are box-plotted per campaign due to the limited number of sky-scans, compared to the high volume of
308 AERONET and in situ observations, centered on the median campaign dates of 21 August 2017, 12 September 2016, and 10
309 October 2018. Log-log spectral quadratic regressions of 4STAR AOD and AAOD were performed, with derivatives taken at
310 500 nm, resulting in EAE_{500} and AAE_{500} , like the procedure performed by Redemann et al. (2006). EAE_{500} (and AAE_{500})
311 were used to extrapolate AOD (and AAOD) from 500 nm to 440 nm, allowing for the calculation of 4STAR SSA at 440 nm.
312 In situ SSA was extrapolated using a spectral linear regression, requiring defined SSA inputs from all three wavelengths
313 (470, 530, 660 nm).

314 After extrapolating to 440 nm, 4STAR and in situ median SSA still show a subseasonal increase of about 0.04 (Fig.
315 6), in agreement with the shorter wavelengths from Fig. 2. This is echoed by the Southern African AERONET stations,
316 which also saw an increase in median SSA of about 0.04 from 23 August to 12 October (the closest 10-day medians to the
317 ORACLES 2017 and 2018 campaign medians), for both the three concurrent campaign years of 2016-2018, as well as for
318 the aerosol climatology of 1995-2025. The AERONET and in situ 10-day SSA medians largely agree on the slope of this
319 increase, barring the very start and end of the ORACLES campaign period. There is good agreement between 4STAR and
320 AERONET SSA medians for ORACLES 2017 and 2018, but the September decrease in SSA shown by 4STAR is not
321 reflected in either the AERONET or in situ datasets. We believe that the southerly location of the ORACLES 2016 campaign
322 impacted 4STAR's September SSA distribution, which is explored further by our spatial analyses.

323 The AERONET station at Windpoort, Namibia (19.37 °S, 15.48 °E) is the greatest contributor of July-October SSA
324 retrievals, supplying 18.5 % of 1995-2025 and 25.3 % of 2016-2018 data, with the latter percentage increasing the station's
325 visibility in Fig. 6b. The second greatest contributor of July-October SSA retrievals is the AERONET station at Mongu Inn,
326 Zambia (15.27 °S, 23.13 °E), supplying 18.0 % of 1995-2025 and 18.7 % of 2016-2018 data, and is well-represented within
327 both panels of Fig. 6 due to its centrality within the latitudinal distribution of AERONET observations. The two AERONET
328 time series are in remarkable agreement with one another, highlighting the ability of the ORACLES 2016-2018 campaigns to
329 reflect a much longer data record. The good agreement between ORACLES and AERONET also suggests that the BBA
330 radiative properties over the Southeast Atlantic remain largely reflective of those emitted from Southern Africa, showcasing
331 how data from ORACLES (and other SEA campaigns) can be relevant outside of its direct study region.

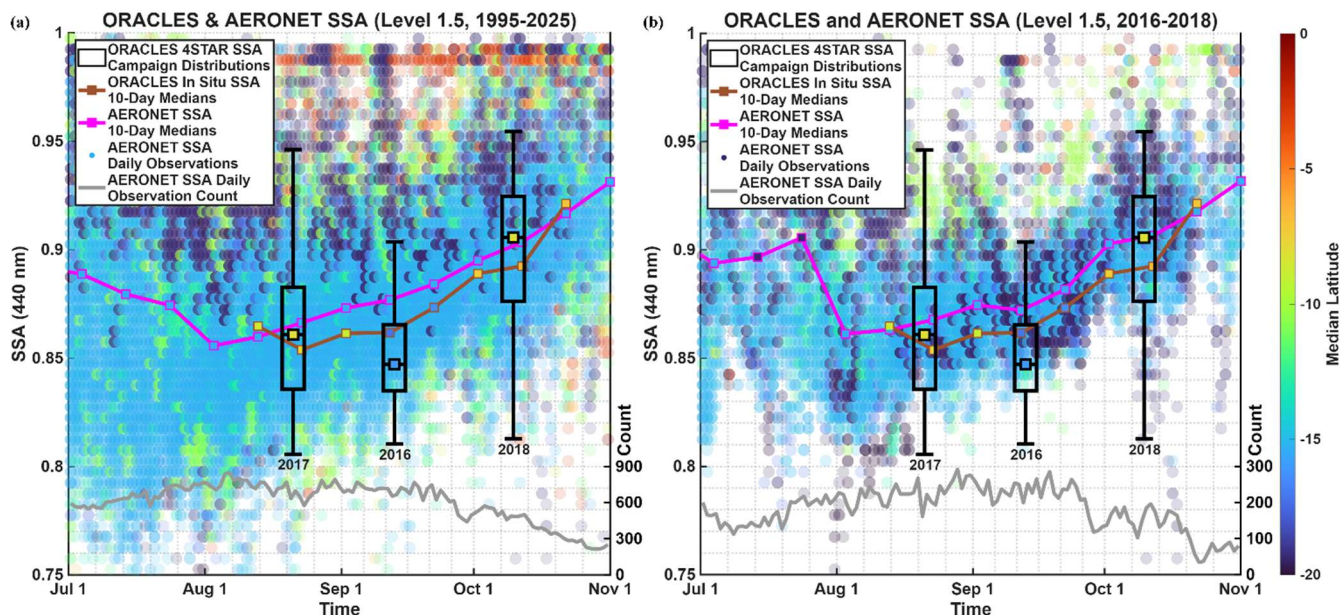


Figure 6: 4STAR and in situ SSA from ORACLES 2016-2018, with AERONET SSA for (a) 1995-2025 and (b) 2016-2018. Black boxes show 4STAR's interquartile range, with square medians, and whiskers extending to the 5th and 95th percentiles. 10-day medians are shown for in situ (brown) and AERONET (pink). Background data are AERONET observations, with opacity weighted by count. The daily count of AERONET observations (gray) is also displayed at the bottom. All other colors represent the median latitude of the observations used to calculate each median SSA value or fill in each cell.

AOD peaks in September for both AERONET and 4STAR at 440 nm (Fig. 7), in agreement with the AOD trend from Fig. 3. Given that AOD is an extensive property, any conclusions drawn from 4STAR are dependent upon its sampling, e.g. when 4STAR operators chose to perform sky-scans and which sky-scans met the QC criteria. Thus, the agreement between 4STAR and AERONET may be somewhat coincidental but also suggests that 4STAR observed a similar distribution of aerosol loadings during sky-scan operation to that obtained by Southern African AERONET stations. The fact that 4STAR observed similar AOD to AERONET (i.e. not observing systematically lower values) implies that aerosol loading was generally conserved during early transport by the Southern African Easterly Jet. A possible mechanism for this is that any aerosol losses may have been compensated by water uptake and hygroscopic growth (Pistone et al., 2021; Pistone et al., 2024) as the aerosols were transported from drier land to moister ocean. Agreement is once again best for ORACLES 2017 and 2018, which had greater sampling of the central smoke plume near 8 °S than the ORACLES 2016 campaign, which may account for the latter's AOD underestimate.

The greatest contributor of July-October AOD measurements is once again the AERONET station at Windpoort, Namibia (19.37 °S, 15.48 °E), supplying 14.7 % of 1995-2025 and 20.3 % of 2016-2018 data, with the latter percentage similarly increasing the station's visibility in Fig. 7b. The AERONET station at Mongu Inn, Zambia (15.27 °S, 23.13 °E) is the second greatest contributor of July-October AOD measurements for 1995-2025, supplying 14.3 % of the aerosol climatology, but is only third greatest for 2016-2018, supplying 14.0 % of the smaller dataset. However, since it is still centrally located within the latitudinal distribution of AERONET observations, it remains well-represented within both

panels of Fig. 7. The second greatest contributor of July-October AOD measurements for 2016-2018 is the AERONET station in Lubango, Namibia (14.96 °S, 13.45 °E), supplying 16.0 % of the smaller dataset, although its contributions are much more muted within the aerosol climatology, at only 8.4 %. The southward trend in African fires can be observed for AERONET AOD greater than 1.0, shifting from near the equator in July, to near 10 °S in August and September, to near 15 °S in October, which is especially apparent in the climatology.

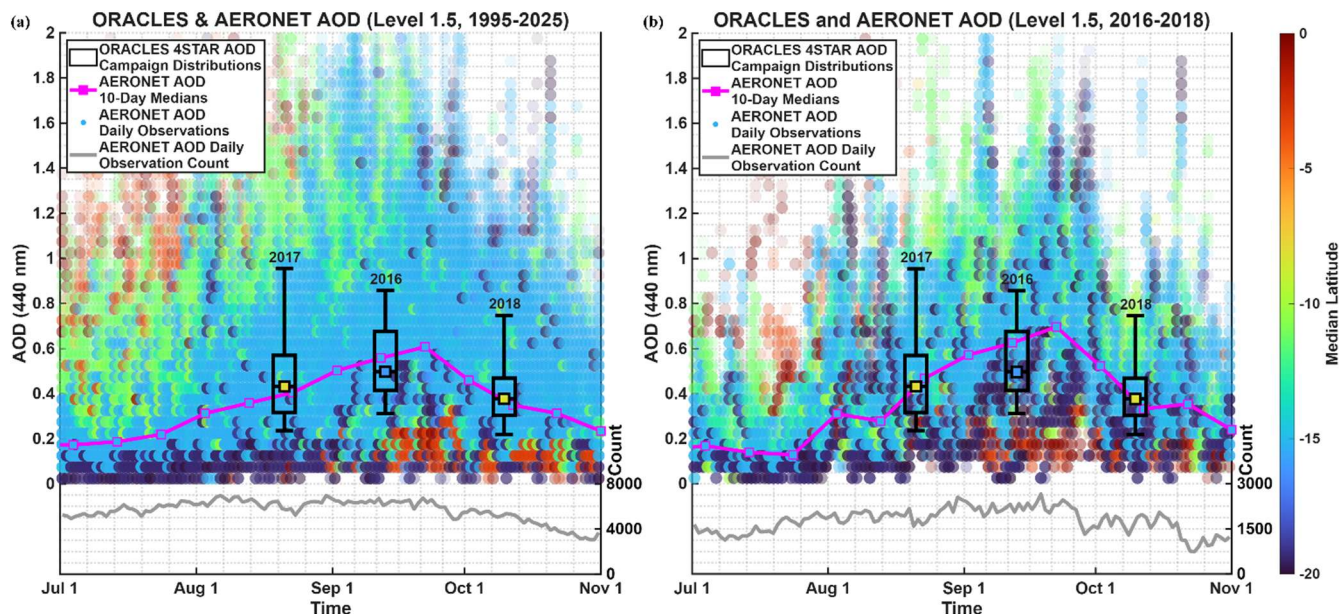


Figure 7: 4STAR AOD from ORACLES 2016-2018, with AERONET AOD for (a) 1995-2025 and (b) 2016-2018. Black boxes show 4STAR's interquartile range, with square medians, and whiskers extending to the 5th and 95th percentiles. AERONET 10-day medians (pink) are also shown. Background data are AERONET observations, with opacity weighted by count. The daily count of AERONET observations (gray) is also displayed at the bottom. All other colors represent the median latitude of the observations used to calculate each median SSA value or fill in each cell.

4.2 Spatial Analyses

By binning SSA by latitude, longitude, and altitude, we can observe its spatial dependences. In situ SSA at 530 nm was chosen for these analyses due to its high volume of data points, as compared to the sparser 4STAR retrievals. For the latitudinal and longitudinal analyses, in situ SSA was binned by 2 °, while for the altitudinal analysis, it was binned by 0.5 km. Each ORACLES campaign was kept separated, so that the subseasonal differences between in situ SSA values at each spatial bin could be observed.

The subseasonal increase in median SSA can be seen for latitudes spanning from 10 °S to 2 °N, with about a 0.03 increase from August to October over that range (Fig. 8). For 10 - 6 °S, all three campaigns display the subseasonal increase, while only the ORACLES 2017 and 2018 campaigns can be observed over the 6 °S to 2 °N range. Over 14 - 10 °S, the SSA medians from August are greater than those from September, not aligning with the subseasonal increase, but August has much fewer samples than the other two campaigns at those latitudes, only containing 6 - 32 % as many datapoints.

378 For ORACLES 2016, median SSA increases by about 0.02 from 20 to 8 °S (excluding south of 20 °S due to
379 possible aerosol recirculations and 8 - 6 °S due to limited datapoints), indicating that ORACLES 2016 observed more
380 aerosol scattering as it approached the central smoke plume near 8 °S. Higher relative humidity near the equator can result in
381 higher SSA values (Ryoo et al., 2021; Dobracki et al., 2023) retrieved by AERONET or 4STAR, but the equatorward
382 increase of in situ SSA cannot solely be attributed to the higher relative humidity, as the PSAP optical block was heated to
383 30 - 50 °C and the relative humidity within the Nephthys had an interquartile range of 6 to 40 % across the three campaigns.
384 This indicates a change in aerosol composition, which is reflective of the equatorward increase in non-BC components noted
385 by the back-trajectories of Dobracki et al. (2025). The aerosol compositional difference is likely due to equatorward fires
386 being smoldering and of woodier materials, generating more organic carbon.

387 Due to the relocation from Namibia to São Tomé, there are significant latitudinal differences between the
388 ORACLES 2016 and the ORACLES 2017-2018 campaigns. South of 14 °S contains 77 % of in situ measurements (and 82
389 % of sky-scans) for ORACLES 2016, but < 1 % of in situ measurements and sky-scans for ORACLES 2017 and 2018.
390 Conversely, north of 8 °S contains 70 - 71 % of in situ measurements (and 46 - 50 % of sky-scans) for ORACLES 2017 and
391 2018, but < 1 % of in situ measurements and sky-scans for ORACLES 2016. We subsampled 4STAR sky-scans from
392 ORACLES 2016 in the overlapping middle region of 14 to 8 °S to determine the effect of the southerly location of that
393 campaign on the SSA retrievals. Doing so yields a median SSA value of 0.86 at 500 nm for that region, which is 0.02 higher
394 than the ORACLES 2016 campaign median. This, combined with the northward increase in ORACLES 2016 in situ SSA
395 medians, suggests that if ORACLES 2016 had been similarly based out of São Tomé and sampled the central smoke plume
396 from the equator, 4STAR and in situ SSA would likely have been higher overall, with greater similarity to AERONET
397 observations and more in alignment with the subseasonal SSA increase.

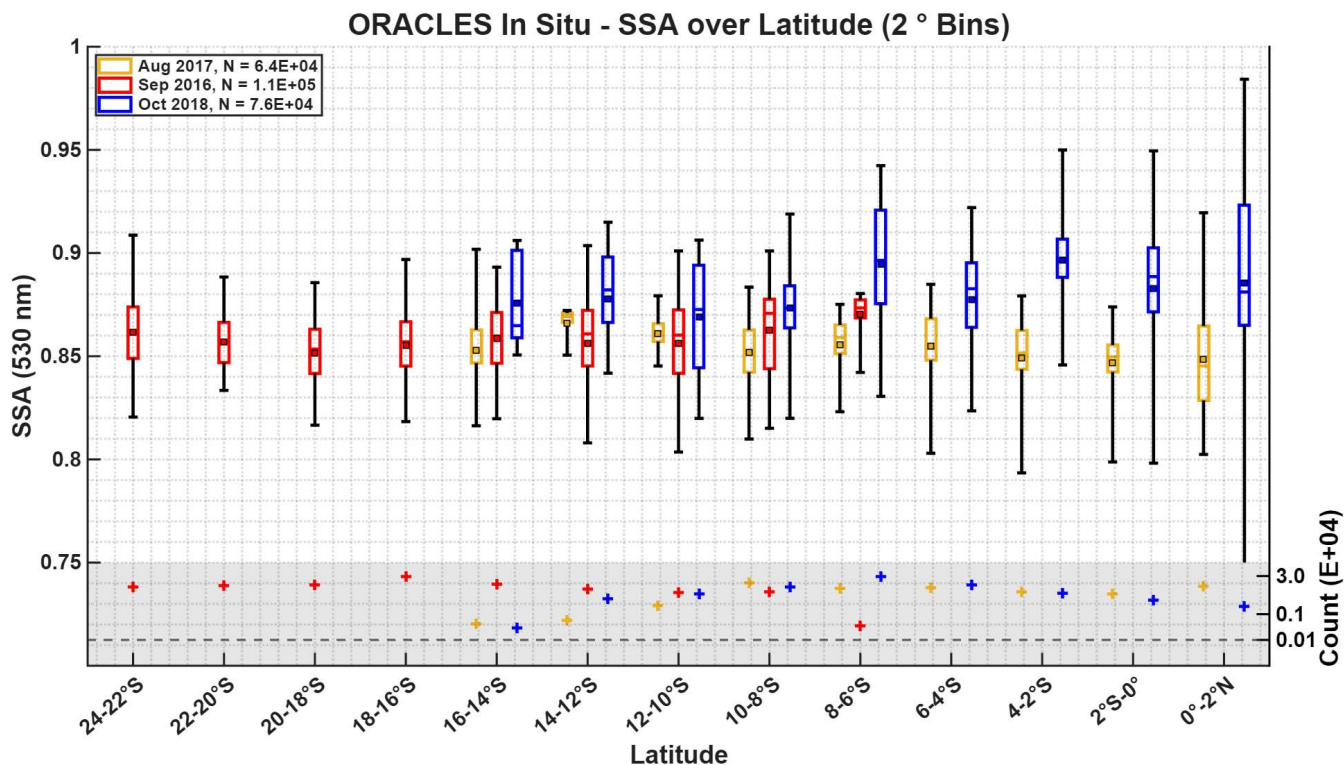


Figure 8: In situ SSA for ORACLES 2016-2018, binned by 2 ° latitude. The ORACLES 2016 (September) campaign is centered, with ORACLES 2017 (August) jittered to its left and ORACLES 2018 (October) to its right. Boxes show the interquartile range, while whiskers extend to the 5th and 95th percentiles. Central lines are medians and squares are means. Observation counts per bin are displayed at the bottom.

The month-to-month increase in median SSA can also be seen for longitudes spanning from 4 °W to 12 °E, with about a 0.03 increase over that range (Fig. 9). For 0 ° to 8 °E, all three campaigns display the subseasonal increase, while only two campaigns can be observed for 4 °W to 0 ° and 8 to 12 °E. The longitudes of 12 - 16 °E were only observed by ORACLES 2016, while 10 - 4 °W were only observed by ORACLES 2017, not allowing for a subseasonal analysis.

In the eastern portion of the basin (16 °E to 2 °W), ORACLES 2016 median SSA is centered near 0.86, with a bin-to-bin variability of about 0.01. Between 10 and 16 °W, ORACLES 2016 median SSA decreases from about 0.84 to 0.80, reflecting a similar SSA decrease found by Fakoya et al. (2025) for aged free tropospheric BBA over the Southeast Atlantic. They attribute this SSA decrease to aerosol aging processes, such as a lensing effect, whereby organic aerosol coatings focus more light on the BC core and increase BBA absorptivity (Taylor et al., 2020), and/or a loss of organic aerosol coating via heterogeneous oxidation and subsequent reduction of BBA scattering (Sedlacek et al., 2022). Over the central portion of the basin (8 °E to 4 °W), ORACLES 2017 median SSA remains almost constant near 0.85, with a bin-to-bin variability of about 0.01. This declines to about 0.83 over 4 - 8 °W, which rebounds to 0.85 for 8 - 16 °W, aligning with the transition from nine-day to eleven-day aged aerosols described by Fakoya et al. (2025). For the east-central portion of the basin (12 °E to 0 °), ORACLES 2018 median SSA decreases westward from about 0.92 to 0.86, then rebounds to 0.88 at 2 - 4 °W. This suggests

that the aerosol aging processes from Fakoya et al. (2025) occurred much further east for ORACLES 2018 than the other two campaigns. This aligns with the southeastward shift of fires in October, allowing more time for aerosol aging processes to occur before arriving in the ORACLES study region over the Southeast Atlantic.

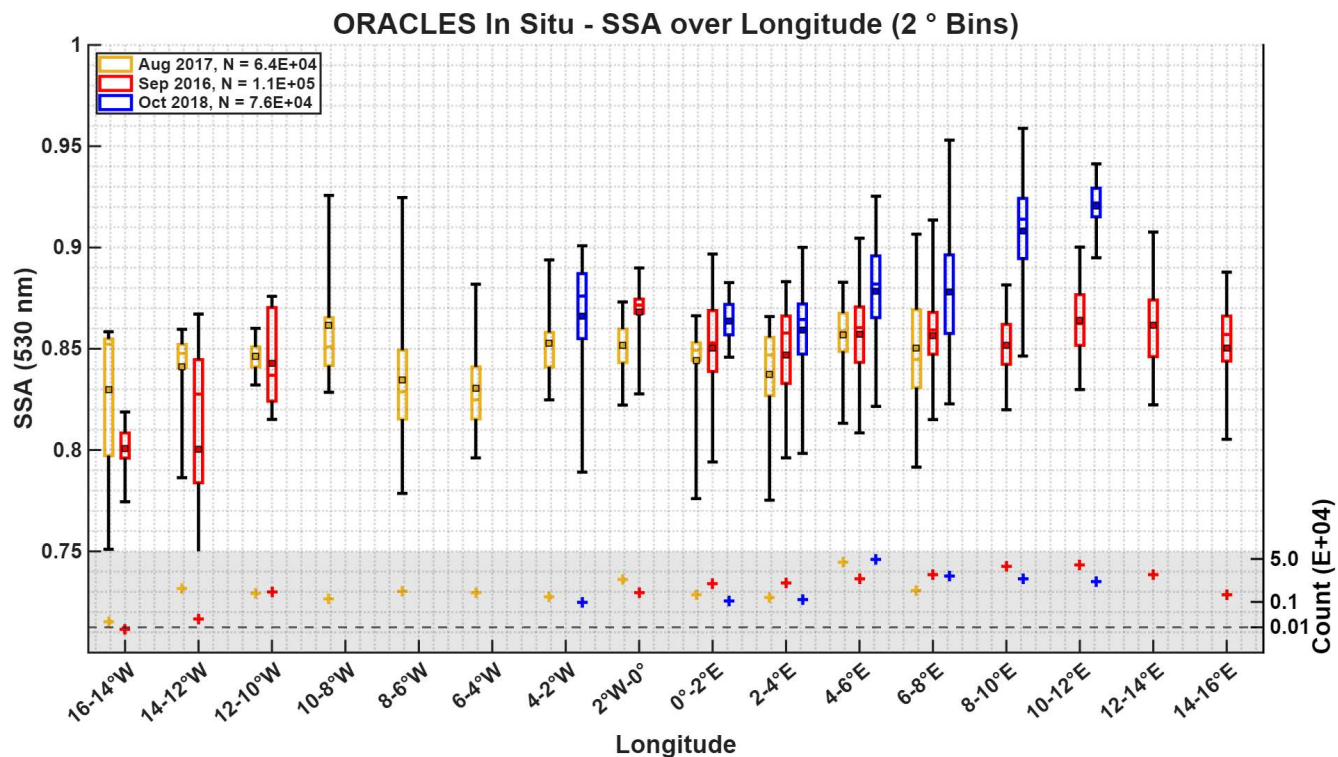


Figure 9: In situ SSA for ORACLES 2016-2018, binned by 2 ° longitude. The ORACLES 2016 (September) campaign is centered, with ORACLES 2017 (August) jittered to its left and ORACLES 2018 (October) to its right. Boxes show the interquartile range, while whiskers extend to the 5th and 95th percentiles. Central lines are medians and squares are means. Observation counts per bin are displayed at the bottom.

SSA generally increases with altitude for all three ORACLES campaigns (Fig. 10), in agreement with Fig. 14 from Redemann et al. (2021), with an average increase of about 0.04 per campaign over the entire free-tropospheric range of 1.5 - 5.5 km. The subseasonal increase in median SSA is strongest in the upper portion of the smoke plume (4 - 5.5 km), with all three campaigns in ascending order and an increase of about 0.05 from August to October over that span. September median SSA is lower than August within the lower portion of the smoke plume (2 - 4 km), but there is still an increase of about 0.03 from August to October. Altitudinal increases in SSA have been attributed to increases in ammonium (NH_3) with altitude (Wu et al., 2020), although not much NH_3 has been measured in the region, estimated to be about 5 % of the mean submicron mass spectrum by Dobracki et al. (2023). The altitudinal increase in SSA is also correlated with a general decrease in the black carbon to organic aerosol ratio with height (Redemann et al., 2021). Another possibility is that stronger winds at higher altitudes result in fresher aerosols aloft, compared to chemically aged aerosols below.

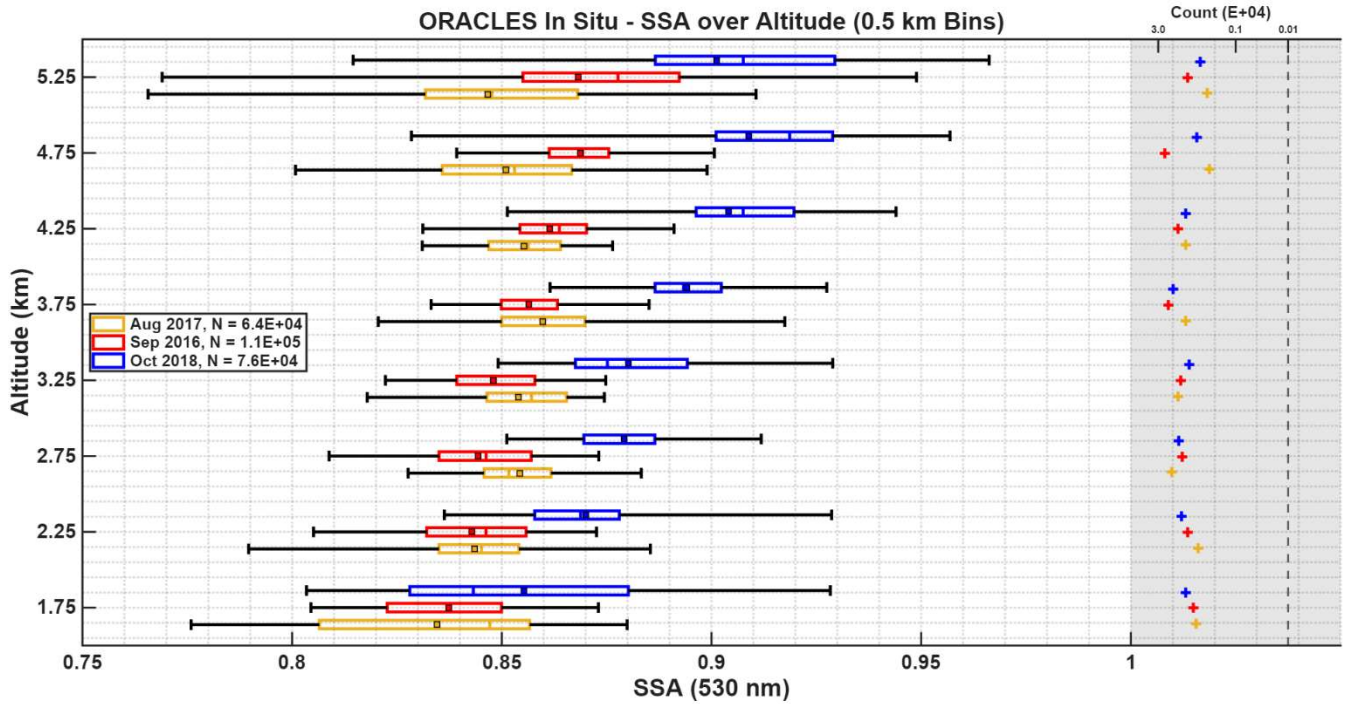


Figure 10: In situ SSA for ORACLES 2016-2018, binned by 0.5 km altitude. Ticks represent the altitudinal center of the bin. The ORACLES 2016 (September) campaign is centered, with ORACLES 2017 (August) jittered below it and ORACLES 2018 (October) above it. Boxes show the interquartile range, while whiskers extend to the 5th and 95th percentiles. Central lines are medians and squares are means. Observation counts per bin are displayed on the right.

5 Conclusions

A subseasonal increase in SSA from 4STAR retrievals and in situ measurements during ORACLES 2016-2018 indicates that biomass burning aerosols over the Southeast Atlantic experience an aerosol brightening process over the course of the BBA emission season. The subseasonal SSA increase is likely to increase critical albedo, which decreases DARE and reduces heating rates in clear and partially cloudy conditions, weakening atmospheric stability. The SSA increase from ORACLES over-ocean observations is also consistent with the near-source observations from Southern African AERONET stations, which have recently reached 30 years of continuous operation, allowing for analysis of a complete aerosol climatology for the first time. EAE and AAE have little subseasonal variation, indicating domination by fine BC aerosols throughout the season, suggesting that the aerosol brightening is due to a change in aerosol composition through the season, rather than a change in aerosol type, although 4STAR did not directly observe the UV wavelengths at which BrC absorption is the strongest. Improving 4STAR's ability to identify BrC absorption via expansion into the UV spectrum is currently complicated by an instrument artifact near 420 nm and stray light scattering at 360 and 380 nm (Mitchell et al., 2025), but could be overcome with the use of hyperspectral GRASP (Generalized Retrieval of Atmosphere and Surface

Properties) code (Román et al., 2018), which is the subject of ongoing work. Regarding the complex refractive indices, the decrease in IRI is relatively greater than the concurrent decrease in RRI, suggesting that the aerosol brightening depends more upon a decrease in aerosol absorptivity, rather than an actual increase in scattering. 4STAR retrievals of AOD and AAOD both peak in September, which is also reflected in the AERONET data. These results are in line with the previously reported pattern of BBA emissions across the source regions.

SSA medians from ORACLES 4STAR retrievals and in situ measurements agree well with those from AERONET, indicating that the BBA radiative properties observed over the Southeast Atlantic are still largely reflective of those emitted from the source region of Southern Africa, with both experiencing a similar subseasonal increase in SSA from aerosol brightening. There is also agreement in AOD medians between ORACLES and AERONET, suggesting that any aerosol losses during transport by the Southern African Easterly Jet may have been compensated by water uptake and hygroscopic growth. The general agreement between these two adjacent regions showcases the relevance of the ORACLES dataset even outside of its direct study area. This is true for both the concurrent campaign years and the full AERONET aerosol climatology, highlighting the ability of ORACLES observations to reflect long term trends.

The SSA and AOD medians retrieved by 4STAR during the ORACLES 2016 campaign are generally lower than the respective AERONET medians. A latitudinal analysis of in situ measurements and subsampling of 4STAR sky-scans indicates that the September decrease in SSA noted by 4STAR is affected by the more southerly sampling location of ORACLES 2016 relative to the other two campaigns, rather than from a temporal evolution, such that ORACLES 2016 would have observed greater scattering if it had similarly targeted the central smoke plume from the equator, bringing the 4STAR SSA medians more in line with the subseasonal increase observed by AERONET. The equatorward increase in SSA also aligns with findings that the source fires within that region are smoldering and of woodier materials. Increased observation of the central smoke plume by ORACLES 2016 would likely have yielded higher aerosol loadings, also bringing AOD into greater agreement with AERONET.

Each campaign observed a westward gradual increase in SSA, followed by a sharper decline, and (in two cases) a subsequent rebound, aligning with late-transport aging processes identified by previous studies, attributed to a lensing effect and/or reduction of organic aerosol coating. This process appears to have occurred much further east for ORACLES 2018, aligning with the southeastward shift of fires in October, allowing more time for aerosol aging processes to occur before arriving to the Southeast Atlantic. SSA generally increases with altitude, in agreement with previous studies, which is attributed to either increasing ammonium, decreasing the black carbon to organic aerosol ratio, or stronger winds keeping fresher aerosols aloft. The subseasonal SSA increase can be seen throughout the free-tropospheric vertical column but is strongest in the upper portion of the smoke plume. 4STAR retrievals and in situ measurements were adept at identifying subseasonal aerosol brightening during the ORACLES campaigns over the Southeast Atlantic, a critical study region that can otherwise be quite difficult to observe.

487 **Data Availability:**

488 NASA ORACLES P-3 4STAR aerosol inversions and in situ absorption measurements are accessible via:
489 https://doi.org/10.5067/ASDC_DAAC/ORACLES_Aerosol_AircraftInSitu_Data_1 (NASA/LARC/SD/ASDC, 2020).
490 AERONET AOD and aerosol inversions are accessible via: <https://aeronet.gsfc.nasa.gov/> (NASA GSFC, 2026).
491 MERRA-2 BC and OC column mass densities (used in a supplementary figure) are accessible via:
492 <https://doi.org/10.5067/FH9A0MLJPC7N> (Global Modeling and Assimilation Office, 2015).
493 Aqua and Terra MODIS-detected vegetation fires (used in a supplementary figure) are accessible via:
494 <https://doi.org/10.5067/FIRMS/MODIS/MCD14DL.NRT.0061> (NASA MODIS Science Data Support Team, 2021).
495 HYSPLIT back-trajectories (used in a supplementary figure) are accessible via: <https://www.ready.noaa.gov/> (NOAA Air
496 Resources Laboratory, 2026).

497 **Author Contributions**

498 All figures were created by LTM, under the guidance of CJF and JR, and with input from KP and SEL. PZ and RW were PIs
499 for ORACLES Mission Science. JR and SEL were PIs for 4STAR during ORACLES 2016 and ORACLES 2017-2018,
500 respectively. KSS was the PI for SSFR during ORACLES. KP and SEL operated the 4STAR instrument aboard the P-3
501 aircraft for all of ORACLES, while CJF also operated during ORACLES 2016. LTM prepared the manuscript with
502 contributions from all co-authors.

503 **Competing Interests**

504 The authors declare that they have no conflict of interest.

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- 513 Abel, S. J., Haywood, J. M., Highwood, E. J., Li, J., and Buseck, P. R.: Evolution of biomass burning aerosol properties
 514 from an agricultural fire in southern Africa, *Geophys. Res. Lett.*, 30, 1783, <https://doi.org/10.1029/2003GL017342>, 2003.
- 515
- 516 Adebisi, A. A., and Zuidema, P.: The role of the southern African easterly jet in modifying the southeast Atlantic aerosol a
 517 and cloud environments, *Q. J. Roy. Meteorol. Soc.*, 142, 1574–1589, <https://doi.org/10.1002/qj.2765>, 2016.
- 518
- 519 Adebisi, A., Zuidema, P., and Abel, S.: The convolution of dynamics and moisture with the presence of shortwave absorbing
 520 aerosols over the southeast Atlantic, *J. Climate*, 28, 1997–2024, <https://doi.org/10.1175/JCLI-D-14-00352.1>, 2015.
- 521
- 522 Alexander, D. T. L., Crozier, P. A., and Anderson, J. R.: Brown carbon spheres in East Asian outflow and their optical
 523 properties, *Science*, 321, 833–836, <https://doi.org/10.1126/science.1155296>, 2008.
- 524
- 525 Anderson, T. L., and Ogren, J. A.: Determining aerosol radiative properties using the TSI 3563 integrating nephelometer,
 526 *Aerosol Sci. Tech.*, 29, 1, 57–69, <https://doi.org/10.1080/02786829808965551>, 1998.
- 527
- 528 Bellouin, N., Quaas, J., Gryspeerdt, E., Kinne, S., Stier, P., Watson-Parris, D., Boucher, O., Carslaw, K. S., Christensen, M.,
 529 Daniau, A.-L., Dufresne, J.-L., Feingold, G., Fiedler, S., Forster, P., Gettelman, A., Haywood, J. M., Lohmann, U.,
 530 Malavelle, F., Mauritsen, T., McCoy, D. T., Myhre, G., Mülmenstädt, J., Neubauer, D., Possner, A., Rugenstein, M., Sato,
 531 Y., Schulz, M., Schwartz, S. E., Sourdeval, O., Storelvmo, T., Toll, V., Winker, D., and Stevens, B.: Bounding global
 532 aerosol radiative forcing of climate change, *Rev. Geophys.*, 58, e2019RG000660, <https://doi.org/10.1029/2019RG000660>,
 533 2020.
- 534
- 535 Bergstrom, R. W., Pilewskie, P., Russell, P. B., Redemann, J., Bond, T. C., Quinn, P. K., and Sierau, B.: Spectral absorption
 536 properties of atmospheric aerosols, *Atmos. Chem. Phys.*, 7, 5937–5943, <https://doi.org/10.5194/acp-7-5937-2007>, 2007.
- 537
- 538 Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher,
 539 B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H.,
 540 Zhang S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz,
 541 J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system:
 542 A scientific assessment, *J. Geophys. Res.-Atmos.*, 118, 5380–5552, <https://doi.org/10.1002/jgrd.50171>, 2013.
- 543
- 544 Bowman, D. M. J. S., Balch, J. K., Artaxo, P., Bond, W. J., Carlson, J. M., Cochrane, M. A., D'Antonio, C. M., DeFries, R.
 545 S., Doyle, J. C., Harrison, S. P., Johnston, F. H., Keeley, J. E., Krawchuk, M. A., Kull, C. A., Marston, J. B., Moritz, M. A.,
 546 Prentice, I. C., Roos, C. I., Scott, A. C., Swetnam, T. W., van der Werf, G. R., and Pyne, S. J.: Fire in the earth system,
 547 *Science*, 324, 481–484, <https://doi.org/10.1126/science.1163886>, 2009.
- 548
- 549 Brown, H., Liu, X., Pokhrel, K., Murphy, S., Lu, Z., Saleh, R., Mielonen, T., Kokkola, H., Bergman, T., Myhre, G., Skeie,
 550 R. B., Watson-Paris, D., Stier, P., Johnson, B., Bellouin, N., Schulz, M., Vakkari, V., Beukes, J. P., van Zyl, P. G., Liu, S.,
 551 and Chand D.: Biomass burning aerosols in most climate models are too absorbing, *Nat. Commun.*, 12, 277,
 552 <https://doi.org/10.1038/s41467-020-20482-9>, 2021.
- 553
- 554 Brown, S. W., Johnson, B. C., Biggar, S. F., Zalewski, E. F., Cooper, J., Hajek, P., Hildum, E., Grant, P., Barnes, R. A., and
 555 Butler, J. J.: Radiometric validation of NASA's Ames Research Center's Sensor Calibration Laboratory, *Appl. Optics*, 44,
 556 6426–6443, <https://doi.org/10.1364/AO.44.006426>, 2005.
- 557
- 558 Chand, D., Wood, R., Anderson, T. L., Satheesh, S. K., and Charlson, R. J.: Satellite-derived direct radiative effect of
 559 aerosols dependent on cloud cover, *Nat. Geosci.*, 2, 181–184, <https://doi.org/10.1038/ngeo437>, 2009.
- 560

Che, H., Stier, P., Gordon, H., Watson-Parris, D., and Deaconu, L.: Cloud adjustments dominate the overall negative aerosol radiative effects of biomass burning aerosols in UKESM1 climate model simulations over the south-eastern Atlantic, *Atmos. Chem. Phys.*, 21, 17–33, <https://doi.org/10.5194/acp-21-17-2021>, 2021.

Choi, M., Lyapustin, A., Schuster, G. L., Go, S., Wang, Y., Korkin, S., Kahn, R., Reid, J. S., Hyer, E. J., Eck, T. F., Chin, M., Diner, D. J., Kalashnikova, O., Dubovik, O., Kim, J., and Moosmüller, H.: Light-absorbing black carbon and brown carbon components of smoke aerosol from DSCOVR EPIC measurements over North America and central Africa, *Atmos. Chem. Phys.*, 24, 10543–10565, <https://doi.org/10.5194/acp-24-10543-2024>, 2024.

Chuvieco, E., Pettinari, M. L., Koutsias, N., Forkel, M., Hantson, S., and Turco, M.: Human and climate drivers of global biomass burning variability, *Sci. Total Environ.*, 779, <https://doi.org/10.1016/j.scitotenv.2021.146361>, 2021.

Cochrane, S. P., Schmidt, K. S., Chen, H., Pilewskie, P., Kittelman, S., Redemann, J., LeBlanc, S., Pistone, K., Kacenelenbogen, M., Segal Rozenhaimer, M., Shinozuka, Y., Flynn, C., Platnick, S., Meyer, K., Ferrare, R., Burton, S., Hostetler, C., Howell, S., Freitag, S., Dobracki, A., and Doherty, S.: Above-cloud aerosol radiative effects based on ORACLES 2016 and ORACLES 2017 aircraft experiments, *Atmos. Meas. Tech.*, 12, 6505–6528, <https://doi.org/10.5194/amt-12-6505-2019>, 2019.

Cochrane, S. P., Schmidt, K. S., Chen, H., Pilewskie, P., Kittelman, S., Redemann, J., LeBlanc, S., Pistone, K., Kacenelenbogen, M., Segal Rozenhaimer, M., Shinozuka, Y., Flynn, C., Dobracki, A., Zuidema, P., Howell, S., Freitag, S., and Doherty, S.: Empirically derived parameterizations of the direct aerosol radiative effect based on ORACLES aircraft observations, *Atmos. Meas. Tech.*, 14, 567–593, <https://doi.org/10.5194/amt-14-567-2021>, 2021.

Cochrane, S. P., Schmidt, K. S., Chen, H., Pilewskie, P., Kittelman, S., Redemann, J., LeBlanc, S., Pistone, K., Segal Rozenhaimer, M., Kacenelenbogen, M., Shinozuka, Y., Flynn, C., Ferrare, R., Burton, S., Hostetler, C., Mallet, M., and Zuidema, P.: Biomass burning aerosol heating rates from the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) 2016 and 2017 experiments, *Atmos. Meas. Tech.*, 15, 61–77, <https://doi.org/10.5194/amt-15-61-2022>, 2022.

Coddington, O., Schmidt, K. S., Pilewskie, P., Gore, W. J., Bergstrom, R. W., Román, M., Redemann, J., Russell, P. B., Liu, J., and Schaaf, C. C.: Aircraft measurements of spectral surface albedo and its consistency with ground-based and spaceborne observations, *J. Geophys. Res.-Atmos.*, 113, D17, <https://doi.org/10.1029/2008JD010089>, 2008.

Collier, S., Zhou, S., Onasch, T. B., Jaffe, D. A., Kleinman, L., Sedlacek III, A. J., Briggs, N. L., Hee, J., Fortner, E., Shilling, J. E., Worsnop, D., Yokelson, R. J., Parworth, C., Ge, X., Xu, J., Butterfield, Z., Chand, D., Dubey, M. K., Pekour, M. S., Springston, S., and Zhang, Q.: Regional influence of aerosol emissions from fires driven by combustion efficiency: Insights from the BBOP campaign, *Environ. Sci. Technol.*, 50, 8613–8622, <https://doi.org/10.1021/acs.est.6b01617>, 2016.

Dang, C., Segal-Rozenhaimer, M., Che, H., Zhang, L., Formenti, P., Taylor, J., Dobracki, A., Purdue, S., Wong, P.-S., Nenes, A., Sedlacek III, A., Coe, H., Redemann, J., Zuidema, P., Howell, S., and Haywood, J.: Biomass burning and marine aerosol processing over the southeast Atlantic Ocean: a TEM single-particle analysis, *Atmos. Chem. Phys.*, 22, 9389–9412, <https://doi.org/10.5194/acp-22-9389-2022>, 2022.

de Graaf, M., Bellouin, N., Tilstra, L. G., Haywood, J., and Stammes, P.: Aerosol direct radiative effect of smoke over clouds over the southeast Atlantic Ocean from 2006 to 2009, *Geophys. Res. Lett.*, 41, 7723–7730, <https://doi.org/10.1002/2014GL061103>, 2014.

Denjean, C., Brito, J., Libois, Q., Mallet, M., Bourrianne, T., Burnet, F., Dupuy, R., Flamant, C., and Knippertz, P.: Unexpected biomass burning aerosol absorption enhancement explained by black carbon mixing state, *Geophys. Res. Lett.*, 47, e2020GL089055, <https://doi.org/10.1029/2020GL089055>, 2020.

Dobracki, A., Zuidema, P., Howell, S. G., Saide, P., Freitag, S., Aiken, A. C., Burton, S. P., Sedlacek III, A. J., Redemann, J., and Wood, R.: An attribution of the low single-scattering albedo of biomass burning aerosol over the southeastern Atlantic, *Atmos. Chem. Phys.*, 23, 4775–4799, <https://doi.org/10.5194/acp-23-4775-2023>, 2023.

Dobracki, A., Lewis, E. R., Sedlacek III, A. J., Tatro, T., Zawadowicz, M. A., and Zuidema, P.: Burning conditions and transportation pathways determine biomass-burning aerosol properties in the Ascension Island marine boundary layer, *Atmos. Chem. Phys.*, 25, 2333–2363, <https://doi.org/10.5194/acp-25-2333-2025>, 2025.

Doherty, S. J., Saide, P. E., Zuidema, P., Shinozuka, Y., Ferrada, G. A., Gordon, H., Mallet, M., Meyer, K., Painemal, D., Howell, S. G., Freitag, S., Dobracki, A., Podolske, J. R., Burton, S. P., Ferrare, R. A., Howes, C., Nabat, P., Carmichael, G. R., da Silva, A., Pistone, K., Chang, I., Gao, L., Wood, R., and Redemann, J.: Modeled and observed properties related to the direct aerosol radiative effect of biomass burning aerosol over the southeastern Atlantic, *Atmos. Chem. Phys.*, 22, 1–46, <https://doi.org/10.5194/acp-22-1-2022>, 2022.

Dubovik, O., and King, M. D.: A flexible inversion algorithm for retrieval of aerosol optical properties from Sun and sky radiance measurements, *J. Geophys. Res.-Atmos.*, 105, 20673–20696, <https://doi.org/10.1029/2000JD900282>, 2000.

Dunagan, S. E., Johnson, R., Zavaleta, J., Russell, P. B., Schmid, B., Flynn, C., Redemann, J., Shinozuka, Y., Livingston, J., Segal-Rosenhaimer, M.: Spectrometer for Sky-Scanning Sun-Tracking Atmospheric Research (4STAR): Instrument Technology, *Remote Sens.*, 5, 3872–3895, <https://doi.org/10.3390/rs5083872>, 2013.

Eck, T. F., Holben, B. N., Reid, J. S., Mukelabai, M. M., Piketh, S. J., Torres, O., Jethva, H. T., Hyer, E. J., Ward, D. E., Dubovik, O., Sinyuk, A., Schafer, J. S., Giles, D. M., Sorokin, M., Smirnov, A., and Slutsker, I.: A seasonal trend of single scattering albedo in southern African biomass-burning particles: Implications for satellite products and estimates of emissions for the world's largest biomass-burning source, *J. Geophys. Res.-Atmos.*, 118, 6414–6432, <https://doi.org/10.1002/jgrd.50500>, 2013.

Fakoya, A. A., Redemann, J., Saide, P. E., Gao, L., Mitchell, L. T., Howes, C., Dobracki, A., Chang, I., Ferrada, G. A., Pistone, K., Leblanc, S. E., Segal-Rosenhaimer, M., Sedlacek III, A. J., Eck, T., Holben, B., Gupta, P., Lind, E., Zuidema, P., Carmichael, G., and Flynn, C. J.: Atmospheric processing and aerosol aging responsible for observed increase in absorptivity of long-range-transported smoke over the southeast Atlantic, *Atmos. Chem. Phys.*, 25, 7879–7902, <https://doi.org/10.5194/acp-25-7879-2025>, 2025.

Formenti, P., D'Anna, B., Flamant, C., Mallet, M., Piketh, S. J., Schepanski, K., Waquet, F., Auriol, F., Brogniez, G., Burnet, F., Chaboureaud, J.-P., Chauvigné, A., Chazette, P., Denjean, C., Desboeufs, K., Doussin, J.-F., Elguindi, N., Feuerstein, S., Gaetani, M., Giorio, C., Klopfer, D., Mallet, M. D., Nabat, P., Monod, A., Solmon, F., Namwoonde, A., Chikwililwa, C., Mushi, R., Welton, E. J., and Holben, B.: The Aerosols, Radiation and Clouds in Southern Africa field campaign in Namibia: Overview, illustrative observations, and way forward, *B. Am. Meteorol. Soc.*, 100, 1277–1298, <https://doi.org/10.1175/bams-d-17-0278.1>, 2019.

Giles, D. M., Sinyuk, A., Sorokin, M. G., Schafer, J. S., Smirnov, A., Slutsker, I., Eck, T. F., Holben, B. N., Lewis, J. R., Campbell, J. R., Welton, E. J., Korkin, S. V., and Lyapustin, A. I.: Advancements in the Aerosol Robotic Network (AERONET) Version 3 database – automated near-real-time quality control algorithm with improved cloud screening for Sun photometer aerosol optical depth (AOD) measurements, *Atmos. Meas. Tech.*, 12, 169–209, <https://doi.org/10.5194/amt-12-169-2019>, 2019.

Global Modeling and Assimilation Office (GMAO): MERRA-2 tavgM_2d_aer_Nx, 2d, Monthly mean, Time-averaged, Single-Level, Assimilation, Aerosol Diagnostics V5.12.4, Goddard Earth Sciences Data and Information Services Center (GES DISC) [data set], <https://doi.org/10.5067/FH9A0MLJPC7N>, 2015.

661 Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., Denier van der Gon, H., Frost, G. J., Heil, A., Kaiser, J. W., Kinne, S.,
662 Klimont, Z., Kloster, S., Lamarque, J.-F., Lioussé, C., Masui, T., Meleux, F., Mieville, A., Ohara, T., Raut, J.-C., Riahi, K.,
663 Schultz, M. G., Smith, S. J., Thompson, A., van Aardenne, J., van der Werf, G. R., and van Vuuren, D. P.: Evolution of
664 anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980–2010 period,
665 *Climatic Change*, 109, 163, <https://doi.org/10.1007/s10584-011-0154-1>, 2011.

666

667 Haywood, J. M., Abel, S. J., Barrett, P. A., Bellouin, N., Blyth, A., Bower, K. N., Brooks, M., Carslaw, K., Che, H., Coe, H.,
668 Cotterell, M. I., Crawford, I., Cui, Z., Davies, N., Dingley, B., Field, P., Formenti, P., Gordon, H., de Graaf, M., Herbert, R.,
669 Johnson, B., Jones, A. C., Langridge, J. M., Malavelle, F., Partridge, D. G., Peers, F., Redemann, J., Stier, P., Szpek, K.,
670 Taylor, J. W., Watson-Parris, D., Wood, R., Wu, H., and Zuidema, P.: The Cloud-Aerosol-Radiation Interaction and
671 Forcing: Year 2017 (CLARIFY-2017) measurement campaign, *Atmos. Chem. Phys.*, 21, 1049–1084,
672 <https://doi.org/10.5194/acp-21-1049-2021>, 2021.

673

674 Holanda, B. A., Pöhlker, M. L., Walter, D., Saturno, J., Sörgel, M., Ditas, J., Ditas, F., Schulz, C., Franco, M. A., Wang, Q.,
675 Donth, T., Artaxo, P., Barbosa, H. M. J., Borrmann, S., Braga, R., Brito, J., Cheng, Y., Dollner, M., Kaiser, J. W., Klimach,
676 T., Knote, C., Krüger, O. O., Fütterer, D., Lavrič, J. V., Ma, N., Machado, L. A. T., Ming, J., Morais, F. G., Paulsen, H.,
677 Sauer, D., Schlager, H., Schneider, J., Su, H., Weinzierl, B., Walser, A., Wendisch, M., Ziereis, H., Zöger, M., Pöschl, U.,
678 Andreae, M. O., and Pöhlker, C.: Influx of African biomass burning aerosol during the Amazonian dry season through
679 layered transatlantic transport of black carbon-rich smoke, *Atmos. Chem. Phys.*, 20, 4757–4785, [https://doi.org/10.5194/acp-](https://doi.org/10.5194/acp-20-4757-2020)
680 [20-4757-2020](https://doi.org/10.5194/acp-20-4757-2020), 2020.

681

682 Holben, B. N., Eck, T. F., Slutsker, I., Tanré, D., Buis, J. P., Setzer, A., Vermote, E., Reagan, J. A., Kaufman, Y. J.,
683 Nakajima, T., Lavenu, F., Jankowiak, I., and Smirnov, A.: AERONET – A federated instrument network and data archive
684 for aerosol characterization, *Remote Sens. Environ.*, 66, 1–16, [https://doi.org/ 10.1016/S0034-4257\(98\)00031-5](https://doi.org/10.1016/S0034-4257(98)00031-5), 1998.

685

686 Holben, B. N., Eck, T. F., Slutsker, I., Smirnov, A., Sinyuk, A., Schafer, J. S., Giles, D. M., and Dubovik, O.: AERONET's
687 version 2.0 quality assurance criteria, *Remote Sensing of Atmosphere and Clouds*, *Proc. SPIE*, 6408, 64080Q,
688 <https://doi.org/10.1117/12.706524>, 2006.

689

690 Keil, A. and Haywood, J. M.: Solar radiative forcing by biomass burning aerosol particles during SAFARI 2000: A case
691 study based on measured aerosol and cloud properties, *J. Geophys. Res.-Atmos.*, 108, 8467,
692 <https://doi.org/10.1029/2002JD002315>, 2003.

693

694 Jen, C. N., Hatch, L. E., Selimovic, V., Yokelson, R. J., Weber, R., Fernandez, A. E., Kreisberg, N. M., Barsanti, K. C., and
695 Goldstein, A. H.: Speciated and total emission factors of particulate organics from burning western US wildland fuels and
696 their dependence on combustion efficiency, *Atmos. Chem. Phys.*, 19, 1013–1026, <https://doi.org/10.5194/acp-19-1013-2019>,
697 2019.

698

699 Jethva, H. T., Torres, O., Ferrare, R. A., Burton, S. P., Cook, A. L., Harper, D. B., Hostetler, C. A., Redemann, J., Kayetha,
700 V., LeBlanc, S., Pistone, K., Mitchell, L., and Flynn, C. J.: Retrieving UV-Vis spectral single-scattering albedo of absorbing
701 aerosols above clouds from synergy of ORACLES airborne and A-train sensors, *Atmos. Meas. Tech.*, 17, 2335–2366,
702 <https://doi.org/10.5194/amt-17-2335-2024>, 2024.

703

704 LeBlanc, S. E., Redemann, R., Flynn, C., Pistone, K., Kacenelenbogen, M., Segal-Rosenheimer, M., Shinozuka, Y.,
705 Dunagan, S., Dahlgren, R. P., Meyer, K., Podolske, J., Howell, S. G., Freitag, S., Small-Griswold, J., Holben, B., Diamond,
706 M., Wood, R., Formenti, P., Piketh, S., Maggs-Kölling, G., Gerber, M., and Namwoonde, A.: Above-cloud aerosol optical
707 depth from airborne observations in the southeast Atlantic, *Atmos. Chem. Phys.*, 20, 1565–1590, [https://doi.org/10.5194/acp-](https://doi.org/10.5194/acp-20-1565-2020)
708 [20-1565-2020](https://doi.org/10.5194/acp-20-1565-2020), 2020.

709

Levin, E. J. T., McMeeking, G. R., Carrico, C. M., Mack, L. E., Kreidenweis, S. M., Wold, C. E., Moosmüller, H., Arnott, W. P., Hao, W. M., Collett Jr., J. L., and Malm, W. C.: Biomass burning smoke aerosol properties measured during Fire Laboratory at Missoula Experiments (FLAME), *J. Geophys. Res.-Atmos.*, 115, D18210, <https://doi.org/10.1029/2009JD013601>, 2010.

Mallet, M., Solmon, F., Nabat, P., Elguindi, N., Waquet, F., Bouniol, D., Sayer, A. M., Meyer, K., Roehrig, R., Michou, M., Zuidema, P., Flamant, C., Redemann, J., and Formenti, P.: Direct and semi-direct radiative forcing of biomass-burning aerosols over the southeast Atlantic (SEA) and its sensitivity to absorbing properties: a regional climate modeling study, *Atmos. Chem. Phys.*, 20, 13191–13216, <https://doi.org/10.5194/acp-20-13191-2020>, 2020.

Meyer, K., Platnick, S., Oreopoulos, L., and Lee, D.: Estimating the direct radiative effect of absorbing aerosols overlying marine boundary layer clouds in the southeast Atlantic using MODIS and CALIOP, *J. Geophys. Res.-Atmos.*, 118, 4801–4815, <https://doi.org/10.1002/jgrd.50449>, 2013.

Mitchell, L. T., Flynn, C. J., Pistone, K., LeBlanc, S. E., Schmidt, K. S., and Redemann, J.: The complete 3-year dataset of 4STAR sky-scans from ORACLES 2016–2018, *Earth Syst. Sci. Data*, 17, 5303–5313, <https://doi.org/10.5194/essd-17-5303-2025>, 2025.

Moosmüller, H., Engelbrecht, J. P., Skiba, M., Frey, G., Chakrabarty, R. K., and Arnott, W. P.: Single scattering albedo of fine mineral dust aerosols controlled by iron concentration, *J. Geophys. Res.-Atmos.*, 117, D11210, <https://doi.org/10.1029/2011JD016909>, 2012.

NASA GSFC: AERONET, <https://aeronet.gsfc.nasa.gov/>, last access: 10 January 2026.

NASA/LARC/SD/ASDC: ORACLES Aerosol Aircraft In Situ Data, NASA Langley Atmospheric Science Data Center DAAC [data set], https://doi.org/10.5067/ASDC_DAAC/ORACLES_Aerosol_AircraftInSitu_Data_1, 2020.

NASA MODIS Science Data Support Team: VIIRS (NOAA-21/JPSS-2) I Band 375 m Active Fire Product NRT (Vector data), NASA LANCE MODIS at the MODAPS [data set], <https://doi.org/10.5067/FIRMS/MODIS/MCD14DL.NRT.0061>, 2021.

NOAA Air Resources Laboratory: HYSPLIT transport model and READY website, <https://www.ready.noaa.gov/>, last access: 05 June 2026.

Pistone, K., Redemann, J., Doherty, S., Zuidema, P., Burton, S., Cairns, B., Cochrane, S., Ferrare, R., Flynn, C., Freitag, S., Howell, S. G., Kacenelenbogen, M., LeBlanc, S., Liu, X., Schmidt, K. S., Sedlacek III, A. J., Segal-Rozenhaimer, M., Shinozuka, Y., Stamnes, S., van Diedenhoven, B., Van Harten, G., and Xu, F.: Intercomparison of biomass burning aerosol optical properties from in situ and remote-sensing instruments in ORACLES-2016, *Atmos. Chem. Phys.*, 19, 9181–9208, <https://doi.org/10.5194/acp-19-9181-2019>, 2019.

Pistone, K., Zuidema, P., Wood, R., Diamond, M., da Silva, A. M., Ferrada, G., Saide, P. E., Ueyama, R., Ryoo, J.-M., Pfister, L., Podolske, J., Noone, D., Bennett, R., Stith, E., Carmichael, G., Redemann, J., Flynn, C., LeBlanc, S., Segal-Rozenhaimer, M., and Shinozuka, Y.: Exploring the elevated water vapor signal associated with the free tropospheric biomass burning plume over the southeast Atlantic Ocean, *Atmos. Chem. Phys.*, 21, 9643–9668, <https://doi.org/10.5194/acp-21-9643-2021>, 2021.

Pistone, K., Wilcox, E. M., Zuidema, P., Giordano, M., Podolske, J., LeBlanc, S. E., Kacenelenbogen, M., Howell, S. G., and Freitag, S.: Vertical structure of a springtime smoky and humid troposphere over the southeast Atlantic from aircraft and reanalysis, *Atmos. Chem. Phys.*, 24, 7983–8005, <https://doi.org/10.5194/acp-24-7983-2024>, 2024.

- Queface, A. J., Piketh, S. J., Eck, T. F., Tsay, S.-C., and Mavume, A. F.: Climatology of aerosol optical properties in Southern Africa, *Atmos. Environ.*, 45, 2910–2921, <https://doi.org/10.1016/j.atmosenv.2011.01.056>, 2011.
- Redemann, J., Zhang, Q., Schmid, B., Russell, P. B., Livingston, J. M., Jonsson, H., and Remer, L. A.: Assessment of MODIS-derived visible and near-IR aerosol optical properties and their spatial variability in the presence of mineral dust, *Geophys. Res. Lett.*, 33, L18814, <https://doi.org/10.1029/2006GL026626>, 2006.
- Redemann, J., Wood, R., Zuidema, P., Doherty, S. J., Luna, B., LeBlanc, S. E., Diamond, M. S., Shinozuka, Y., Chang, I. Y., Ueyama, R., Pfister, L., Ryoo, J. M., Dobracki, A. N., da Silva, A. M., Longo, K. M., Kacenelenbogen, M. S., Flynn, C., Pistone, K., Knox, N. M., Piketh, S. J., Haywood, J., Formenti, P., Mallet, M., Stier, P., Ackerman, A. S., Bauer, S. E., Fridlind, A. M., Carmichael, G. R., Saide, P. E., Ferrada, G. A., Howell, S. G., Cairns, B., Holben, B. N., Knobelspiesse, K. D., Tanelli, S., L’Ecuyer, T. S., Dzambo, A. M., Sy, O. O., McFarquhar, G. M., Poellot, M. R., Gupta, S., O’Brien, J. R., Nenes, A., Kacarab, M., Wong, J. P. S., Small-Griswold, J. D., Thornhill, K. L., Noone, D., Podolske, J. R., Schmidt, K. S., Pilewskie, P., Chen, H., Cochrane, S. P., Sedlacek, A. J., Lang, T. J., Stith, E., Segal-Rosenhaimer, M., Ferrare, R. A., Burton, S. P., Hostetler, C. A., Diner, D. J., Seidel, F. C., Platnick, S. E., Myers, J. S., Meyer, K. G., Spangenberg, D. A., Maring, H., and Gao, L.: An overview of the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) project: aerosol-cloud-radiation interactions in the southeast Atlantic basin, *Atmos. Chem. Phys.*, 21, 1507–1563, <https://doi.org/10.5194/acp-21-1507-2021>, 2021.
- Rolph, G., Stein, A., and Stunder, B.: Real-time Environmental Applications and Display sYstem: READY, *Environ. Modell. Software*, 95, 210–228, <https://doi.org/10.1016/j.envsoft.2017.06.025>, 2017.
- Román, R., Benavent-Oltra, J. A., Casquero-Vera, J. A., Lopatin, A., Cazorla, A., Lyamani, H., Denjean, C., Fuertes, D., Pérez-Ramírez, D., Torres, B., Toledano, C., Dubovik, O., Cachorro, V. E., de Frutos, A. M., Olmo, F. J., and Alados-Arboledas, L.: Retrieval of aerosol profiles combining sunphotometer and ceilometer measurements in GRASP code, *Atmos. Res.*, 204, 161–177, <https://doi.org/10.1016/j.atmosres.2018.01.021>, 2018.
- Russell, P. B., Bergstrom, R. W., Shinozuka, Y., Clarke, A. D., DeCarlo, P. F., Jimenez, J. L., Livingston, J. M., Redemann, J., Dubovik, O., and Strawa, A.: Absorption Angstrom Exponent in AERONET and related data as an indicator of aerosol composition, *Atmos. Chem. Phys.*, 10, 1155–1169, <https://doi.org/10.5194/acp-10-1155-2010>, 2010.
- Ryoo, J.-M., Pfister, L., Ueyama, R., Zuidema, P., Wood, R., Chang, I., and Redemann, J.: A meteorological overview of the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) campaign over the southeastern Atlantic during 2016–2018: Part 1 – Climatology, *Atmos. Chem. Phys.*, 21, 16689–16707, <https://doi.org/10.5194/acp-21-16689-2021>, 2021.
- Sakaeda, N., Wood, R., and Rasch, P. J.: Direct and semidirect aerosol effects of southern African biomass burning aerosol, *J. Geophys. Res.-Atmos.*, 116, D12, <https://doi.org/10.1029/2010JD015540>, 2011.
- Schmid, B., and Wehrli, C.: Comparison of Sun photometer calibration by use of the Langley technique and the standard lamp. *Appl. Optics*, 34, 4500–4512, <https://doi.org/10.1364/AO.34.004500>, 1995.
- Schmidt, S. and Pilewskie, P.: Airborne measurements of spectral shortwave radiation in cloud and aerosol remote sensing and energy budget studies, in: *Light Scattering Reviews*, Vol. 6, edited by: Kokhanovsky, A. A., Springer, Berlin, Heidelberg, Germany, 239–288, https://doi.org/10.1007/978-3-642-15531-4_6, 2012.
- Schmidt, K. S., Wendisch, M., and Kindel, B.: Airborne solar radiation sensors, in: *Springer Handbook of Atmospheric Measurements*, edited by: Foken, T., Springer, Cham, Switzerland, 1131–1150, https://doi.org/10.1007/978-3-030-52171-4_40, 2021.

- Schuster, G. L., Dubovik, O., and Holben, B. N.: Angstrom exponent and bimodal aerosol size distributions, *J. Geophys. Res.-Atmos.*, 111, D07207, <https://doi.org/10.1029/2005JD006328>, 2006.
- Sedlacek, A. J., III, Lewis, E. R., Onasch, T. B., Zuidema, P., Redemann, J., Jaffe, D., and Kleinman, L. I.: Using the Black Carbon Particle Mixing State to Characterize the Lifecycle of Biomass Burning Aerosols, *Environ. Sci. Technol.*, 56, 14315–14325, <https://doi.org/10.1021/acs.est.2c03851>, 2022.
- Segal-Rosenheimer, M., Russell, P. B., Schmid, B., Redemann, J., Livingston, J. M., Flynn, C. J., Johnson, R. R., Dunagan, S. E., Shinozuka, Y., Herman, J., Cede, A., Abuhassan, N., Comstock, J. M., Hubbe, J. M., Zelenyuk, A., and Wilson, J.: Tracking elevated pollution layers with a newly developed hyperspectral sun/sky spectrometer (4STAR): Results from the TCAP 2012 and 2013 campaigns. *J. Geophys. Res.-Atmos.*, 119, 5, 2611–2628, <https://doi.org/10.1002/2013JD020884>, 2014.
- Sinyuk, A., Holben, B. N., Eck, T. F., Giles, D. M., Slutsker, I., Korkin, S., Schafer, J. S., Smirnov, A., Sorokin, M., and Lyapustin, A.: The AERONET Version 3 aerosol retrieval algorithm, associated uncertainties and comparisons to Version 2, *Atmos. Meas. Tech.*, 13, 3375–3411, <https://doi.org/10.5194/amt-13-3375-2020>, 2020.
- Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D., and Ngan, F.: NOAA's HYSPLIT atmospheric transport and dispersion modeling system, *Bull. Am. Meteorol. Soc.*, 96, 2059–2077, <http://doi.org/10.1175/BAMS-D-14-00110.1>, 2015.
- Stier, P., Schutgens, N. A. J., Bellouin, N., Bian, H., Boucher, O., Chin, M., Ghan, S., Huneus, N., Kinne, S., Lin, G., Ma, X., Myhre, G., Penner, J. E., Randles, C. A., Samset, B., Schulz, M., Takemura, T., Yu, F., Yu, H., and Zhou, C.: Host model uncertainties in aerosol radiative forcing estimates: results from the AeroCom Prescribed intercomparison study, *Atmos. Chem. Phys.*, 13, 3245–3270, <https://doi.org/10.5194/acp-13-3245-2013>, 2013.
- Takemura, T., Nakajima, T., Dubovik, O., Holben, B. N., and Kinne, S.: Single-scattering albedo and radiative forcing of various aerosol species with a global three-dimensional model, *J. Climate*, 15, 333–352, [https://doi.org/10.1175/1520-0442\(2002\)015<0333:SSAARF>2.0.CO;2](https://doi.org/10.1175/1520-0442(2002)015<0333:SSAARF>2.0.CO;2), 2002.
- Tatro, T., and Zuidema, P.: More biomass burning aerosol is being advected westward over the southern tropical Atlantic since 2003, *Sci. Total Environ.*, 965, <https://doi.org/10.1016/j.scitotenv.2025.178506>, 2025.
- Taylor, J. W., Wu, H., Szpek, K., Bower, K., Crawford, I., Flynn, M. J., Williams, P. I., Dorsey, J., Langridge, J. M., Cotterell, M. I., Fox, C., Davies, N. W., Haywood, J. M., and Coe, H.: Absorption closure in highly aged biomass burning smoke, *Atmos. Chem. Phys.*, 20, 11201–11221, <https://doi.org/10.5194/acp-20-11201-2020>, 2020.
- van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), *Atmos. Chem. Phys.*, 10, 11707–11735, <https://doi.org/10.5194/acp-10-11707-2010>, 2010.
- Virkkula, A.: Correction of the calibration of the 3-wavelength Particle Soot Absorption Photometer (3 λ PSAP), *Aerosol Sci. Tech.*, 44, 8, 706–712, <https://doi.org/10.1080/02786826.2010.482110>, 2010.
- Waquet, F., Peers, F., Ducos, F., Goloub, P., Platnick, S., Riedi, J., Tanré, D., and Thieuleux, F.: Global analysis of aerosol properties above clouds, *Geophys. Res. Lett.*, 40, 5809–5814, <https://doi.org/10.1002/2013GL057482>, 2013.
- Wilcox, E. M.: Direct and semi-direct radiative forcing of smoke aerosols over clouds, *Atmos. Chem. Phys.*, 12, 139–149, <https://doi.org/10.5194/acp-12-139-2012>, 2012.

860 Wu, H., Taylor, J. W., Szpek, K., Langridge, J. M., Williams, P. I., Flynn, M., Allan, J. D., Abel, S. J., Pitt, J., Cotterell, M.
861 I., Fox, C., Davies, N. W., Haywood, J., and Coe, H.: Vertical variability of the properties of highly aged biomass burning
862 aerosol transported over the southeast Atlantic during CLARIFY-2017, *Atmos. Chem. Phys.*, 20, 12697–12719,
863 <https://doi.org/10.5194/acp-20-12697-2020>, 2020.

864
865 Zhang, A., Wang, Y., Zhang, Y., Weber, R. J., Song, Y., Ke, Z., and Zou, Y.: Modeling the global radiative effect of brown
866 carbon: a potentially larger heating source in the tropical free troposphere than black carbon, *Atmos. Chem. Phys.*, 20, 1901–
867 1920, <https://doi.org/10.5194/acp-20-1901-2020>, 2020.

868
869 Zhang, L., Segal-Rozenhaimer, M., Che, H., Dang, C., Sedlacek III, A. J., Lewis, E. R., Dobracki, A., Wong, J. P. S.,
870 Formenti, P., Howell, S. G., and Nenes, A.: Light absorption by brown carbon over the South-East Atlantic Ocean, *Atmos.*
871 *Chem. Phys.*, 22, 9199–9213, <https://doi.org/10.5194/acp-22-9199-2022>, 2022.

872
873 Zuidema, P., Redemann, J., Haywood, J., Wood, R., Piketh, S. J., Hipondoka, M., and Formenti, P.: Smoke and Clouds
874 above the Southeast Atlantic: Upcoming Field Campaigns Probe Absorbing Aerosol's Impact on Climate, *B. Am. Meteorol.*
875 *Soc.*, 97, 1131–1135, <https://doi.org/10.1175/BAMS-D-15-00082.1>, 2016.

876
877 Zuidema, P., Sedlacek III, A. J., Flynn, C., Springston, S., Delgadillo, R., Zhang, J., Aiken, A. C., Koontz, A., and
878 Muradyan, P.: The Ascension Island boundary layer in the remote Southeast Atlantic is often smoky, *Geophys. Res. Lett.*,
879 45, 4456–4465, <https://doi.org/10.1002/2017GL076926>, 2018.