Geophysical Research Letters

RESEARCH LETTER
10.1002/2017GL076926

The Ascension Island Boundary Layer in the Remote Southeast Atlantic is Often Smoky

Paquita Zuidema1, Arthur J. Sedlacek III2, Connor Flynn3, Stephen Springston3, Rodrigo Delgadillo1, Jianhao Zhang1, Allison C. Aiken4, Annette Koontz3, and Paytsar Muradyan5

1Rosenstiel School of Marine and Atmospheric Sciences, University of Miami, Miami, FL, USA, 2Brookhaven National Laboratory, Brookhaven, NY, USA, 3Pacific Northwest National Laboratory, Richland, WA, USA, 4Los Alamos National Laboratory, Los Alamos, NM, USA, 5Argonne National Laboratory, Argonne, IL, USA

Abstract Observations from June to October 2016, from a surface-based ARM Mobile Facility deployment on Ascension Island (8°S, 14.5°W) indicate that refractory black carbon (rBC) is almost always present within the boundary layer. The rBC mass concentrations, light absorption coefficients, and cloud condensation nuclei concentrations vary in concert and synoptically, peaking in August. Light absorption coefficients at three visible wavelengths as a function of rBC mass are approximately double that calculated from black carbon in lab studies. A spectrally-flat absorption angstrom exponent suggests most of the light absorption is from lens-coated black carbon. The single-scattering-albedo increases systematically from August to October in both 2016 and 2017, with monthly means of 0.78 ± 0.02 (August), 0.81 ± 0.03 (September), and 0.83 ± 0.03 (October) at the green wavelength. Boundary layer aerosol loadings are only loosely correlated with total aerosol optical depth, with smoke more likely to be present in the boundary layer earlier in the biomass burning season, evolving to smoke predominantly present above the cloud layers in September–October, typically resting upon the cloud top inversion. The time period with the campaign-maximum near-surface light absorption and column aerosol optical depth, on 13–16 August 2016, is investigated further. Backtrajectories that indicate more direct boundary layer transport westward from the African continent is central to explaining the elevated surface aerosol loadings.

Plain Language Summary First findings from the remote Ascension Island midway between Africa and South America in the Atlantic Ocean indicate that smoke is present much more often near the surface than has been previously thought. The new measurements from a 17-month-long campaign suggest that August is the smokiest month near the surface. The smoke includes other aerosols besides black carbon, and is most absorptive of sunlight in June and least in October. The smoke is more present near the surface earlier in the biomass burning season, or June, while later on toward September and October, more of the smoke resides above the cloud layer. This has implications for which aerosol-cloud microphysical and radiative interactions are dominant when. The campaign-maximum aerosol loading event is investigated further and attributed to an unusual direct westward flow from the continental African fire sources at low altitudes.

1. Introduction

Space-based observations provided the first suggestion that the remote Southern Hemisphere was not pristine, despite a lack of major population centers at the time (Fishman et al., 1991). Since then, it has become well-established that shortwave-absorbing aerosol emanating from biomass burning in continental Africa advects westward over the southern Atlantic for approximately one third of the year, from June to October (Adebiyi et al., 2015; Garstang et al., 1996; Kaufman et al., 2003). This smoke coincides with an equally expansive stratocumulus deck that contributes a significant top-of-atmosphere net cloud radiative cooling (e.g., Lin et al., 2010). The absorption of sunlight by the smoke above a bright underlying surface can contribute a climate warming, though its value is sensitive to the smoke's single-scattering albedo (SSA; Abel et al., 2005; Chand et al., 2009; Russell et al., 1997; Seidel & Popp, 2012) among other factors. This is in turn a function of aerosol composition, age, and size (Bond et al., 2013; Eck et al., 2013; Haywood et al., 2003; Leahy et al., 2007; Magi et al., 2008; Waquet et al., 2013).
The coincidence of smoke and low-level cloud furthermore encourages small-scale aerosol-cloud-radiation interactions that can potentially produce additional effects on regional climate and precipitation patterns (Ramanathan & Carmichael, 2008; Randles & Ramaswamy, 2010; Sakaeda et al., 2011). These interactions are still poorly understood but to first order will depend crucially on whether the shortwave-absorbing aerosols and cloud can physically interact (Ackerman et al., 2000; Costantino & Bréon, 2013; Johnson et al., 2004; Johnson, 2005; Koch & Del Genio, 2010; Lu et al., 2018; Painemal et al., 2014; Yamaguchi et al., 2015; Wilcox et al., 2016; Zhou et al., 2017). Space-based lidar provides ever-improving estimates of the aerosol-cloud vertical structure (Liu et al., 2015; Rajapakse et al., 2017), but clouds will always obscure a space-based lidar’s ability to infer biomass burning aerosol presence and properties in the boundary layer. Prior to 2016, only one robust in situ aircraft profile had determined that aerosol can be present at elevated levels in the remote southeast Atlantic boundary layer (Haywood et al., 2003).

An important step toward acquiring new knowledge occurred with the deployment of the Department of Energy Atmospheric Radiation Measurement Mobile Facility 1 to Ascension Island (8°S and 14.5°W) for 17 months, from 1 June 2016 to 31 October 2017, as part of the Layered Atlantic Smoke Interactions with Clouds (LASIC; Zuidema et al., 2015) campaign. The island is located midway between continental Africa and South America, within warm oceans that encourage deeper boundary layers and underlies the primary smoke plume emanating off of continental southern Africa in the boreal fall (Adebiyi & Zuidema, 2016). Surface-based in situ measurements of aerosol are complemented by micropulse lidar observations of the aerosol vertical structure. Here we document an important early finding, namely, that high loadings of black carbon and shortwave absorptive aerosols are common in the boundary layer during June to August. These vary with the month, with indications of more organic aerosol in June. The SSA increases from August to October, with monthly-mean values of 0.78 ± 0.02, 0.81 ± 0.03 to 0.83 ± 0.03 at the green wavelength, respectively averaged over both 2016 and 2017. More of the total column aerosol is in the boundary layer early in the biomass burning aerosol season, migrating to relatively more aerosol above the cloudy boundary layer in September–October. The time period with the most near-surface shortwave absorption of the entire 17-month-long campaign, occurring on 13–16 August 2016, is investigated further.

### 2. Surface-Based Aerosol Measurements

Measurements from the first biomass burning season, spanning June–October 2016, include time series of the refractory black carbon (rBC) mass concentrations derived from the single-particle soot photometer (SP2), the shortwave absorption at three wavelengths (blue = 464 nm, green = 529 nm, and red = 648 nm), the cloud condensation nuclei (CCN) concentrations at 0.1%, 0.2%, and 0.4% supersaturations, and the condensation particle concentrations (particle diameters >10 nm) and carbon monoxide (Figure 1). The rBC mass concentrations already regularly exceed 10 ng/m³ by June. The variability is predominantly synoptic, with smokier and cleaner time periods able to alternate within a few days of each other. August is the smokiest month depicted, with a monthly-mean rBC mass concentration reaching almost 500 ng/m³. The maximum rBC exceeded 1,700 ng/m³ on 13 August 2016, followed by another local maximum on 30 August. The peaks in the near-surface values are comparable to those measured closer to the African coast between 3 and 6 km altitude in September 2016 by a NASA P-3 research plane. A strong subsequent decline in September occurs to much lower and even occasionally zero concentrations of black carbon. Nevertheless, even in November rBC mass values still exceeded 3 ng/m³ (the SP2 detection limit) 90% of the time, although overall the monthly-mean value was much lower at 32 ng/m³. Few island sources exist for the black carbon, with the instrumentation located on the windward side of the island upwind from the site generator. Details of the aerosol light absorption variability with time track that of the rBC. Most of the aerosol can be activated into CCN by supersaturations of 0.2%, more prominently so in June, indicating the aerosol’s potential to modify clouds microphysically. A supersaturation of 0.2% is readily achieved in the marine environment (e.g., Wood et al., 2012). At 0.4% supersaturation, all or nearly so of the condensation particles with diameters >10 nm are activated. Carbon monoxide (CO) values indicate that clean background conditions representative of the atmosphere above the southern high-latitude ocean, can also occur, sometimes within only a few days of a heavy smoke event. Also evident is that the CO:particle count ratio increases over time; the CO:rBC ratio is also lower in June than in August–September, implying that either the aerosol in June is more aged (Heald et al., 2004) or stems from more actively flaming sources producing less CO compared to smoldering fires (e.g., Liu et al., 2014).
Figure 1. The 1 June to 31 October 2016 time series of (a) single-particle soot photometer (SP2)-derived refractory black carbon mass concentrations. Monthly 10th, 25th, 50th, 75th, 90th percentiles are indicated, with a dotted line connecting monthly-mean values. (b) Particle soot absorption photometer (PSAP) aerosol light absorption coefficients at three wavelengths (blue: 464 nm, green: 529 nm, and red: 648 nm) as an average of the Virkkula (2010) and Ogren (2010) corrections (see supporting information for more detail). The inset indicates the relative frequency distribution of the blue-red absorption angstrom exponent, only calculated when the blue nephelometer-derived scattering $>10 \, \text{Mm}^{-1}$. (c) Cloud condensation concentrations (CCN) at 0.1%, 0.2%, and 0.4% supersaturations (data from 15 September to 1 November are missing). Inset indicates daily averaged CCN versus refractory black carbon mass concentrations. (d) Condensation particle concentrations (black; minimum particle diameter of 10 nm) and carbon monoxide (red). See supporting information for more data description of all figures.
Aerosols accompanying the black carbon also contribute to the radiative and microphysical environment of the remote southeast Atlantic boundary layer. This is evident in mass absorption cross sections (aerosol light absorption as a function of rBC mass loading) that are consistently 1.5 to 2.5 times that of nascent (or uncoated) black carbon (Figures 2a–2c; Bond et al., 2013), reaching similar values to urban industrial pollution (e.g., Peng et al., 2016). The carbonaceous aerosol absorbs more sunlight in June than in July–September, (Figures 2a–2c). A lens coating of the rBC is one way to reconcile these mass absorption cross-sections with the spectrally flat June–October mean blue-red absorption angstrom exponent (AAE) of 1.08 (Figure 1b,
Figure 3. (a) The 1 June to 31 October 2016 light extinction time series (daily averaged, 529 nm; as the sum of nephelometer-derived scattering and Particle soot absorption photometer-derived absorption) and 500 nm AERONET Sun photometer aerosol optical depths (Version 3, Level 1.5, airport site). Inset is a scatterplot of the same data subsampled for coincidence. (b) same as (a) but for 1 June to 31 October, 2017.

inset; the mean AAE value is highest in June at 1.16. Hygroscopicity of accompanying aerosol (Perring et al., 2017) can also explain why the CCN maximum (>1,100 cm⁻³ on 24 June 2016), precedes the August rBC maximum (Figure 1c).

The SSA, derived when scattering coefficients exceed 10 ng/m³ only, increase from August to October (Figure 2d). A similar SSA trend is evident in 2017 (Figure 2e), indicating that it is a robust feature. Averaged over the two years, the SSA increases from 0.78 ± 0.02 in August to 0.81 ± 0.03 in September and 0.83 ± 0.03 in October, at 532 nm wavelength. The reproducibility of the trend across the two years supports the assumption of the contemporary multiyear NASA ORACLES (Observations of Aerosols above Clouds and their interactions) aircraft campaign that sampling different months in different years can provide insight into the seasonal cycle in aerosol properties (see Zuidema, Redemann, et al., 2016, for more description of aircraft campaigns overlapping with LASIC). The SSA values are least variable in August, when the boundary layer aerosol loadings are also the highest. The SSA values are based on a quality-controlled data set, although the contributing measurements occur at nonconstant relative humidities; see supporting information for more discussion. The mean August–September SSA values are lower than those previously reported based on column closure studies over southern Africa by Leahy et al., 2007 (0.80 ± 0.02 versus 0.85 ± 0.02). While limitations with filter-based measurements of aerosol light absorption cannot be ignored (Coen et al., 2010; Virkkula et al., 2005; Virkkula, 2010), reasonable physical explanations for the increased absorption at the Ascension Island surface relative to southern Africa also exist. Coatings of nonrefractory material on the rBC can absorb additional sunlight depending on the coating thickness (Lack et al., 2009; Zhang et al., 2008), with absorption increasing at higher relative humidities (e.g., Brem et al., 2012). The composition of the accompanying, shortwave-absorbing aerosol can also differ (Eck et al., 2013; Liu et al., 2014). Surface SSA values may will also differ from the column average. In the lowest 1.5 km, relative humidities readily reach 100%, and hygroscopic particle growth will increase scattering (see, e.g., Figure 5c). Leahy et al. (2007) similarly found that the column
average SSA values exceeded the aircraft-derived values in the middle troposphere by 0.02. Thus, the Ascension near-surface filter-based SSA values can be reasonable, though further research is required to place this conclusion on firmer ground.

3. Relationship of the Surface-Based Aerosol Concentrations to Total Aerosol

Daily-mean aerosol optical depths over the June–October time periods occasionally exceed 0.4, with a maximum of almost 0.7 on 14 August 2016, although mean values over the June–October time periods of both years were not substantial at approximately 0.2 (Figure 3). More interesting is the lack of a consistent correlation between the surface-based light extinction and the total column aerosol optical depth. Rather, separate regimes are evident, with the boundary layer aerosol more likely to dominate the aerosol optical depth in June–August (most clearly in June, when a smokier boundary layer can coexist with a relatively low aerosol optical depth), while by late August through October, higher aerosol optical depths are more likely to cooccur with reduced rBC mass loadings at the surface, most noticeable in September 2017. The lower atmosphere is annually the most stable in September (Richter & Mechoso, 2004; Zuidema, Chang, et al., 2016), discouraging the entrainment of smoke from above into the cloud layer, with stronger zonal winds between 3 and 5 km more effectively transporting carbonaceous aerosol over longer ranges in September–October (Adebiyi & Zuidema, 2016).

4. The 13–16 August 2016 Case Study

Figure 4 highlights the time period with the highest rBC loadings, light absorption, and aerosol optical depth of the two biomass burning seasons. On 13 August 2016, closed-cell cumulus clusters with cores reaching almost 2 km generate upper level stratiform cloud, with lower local orographically-lifted cloud at the AMF1 site fully attenuating the lidar signal. The following day is characterized by more suppressed wind-aligned shallow cumuli that allowed the micropulse lidar to more fully probe the atmosphere. Smoke is present to above 3 km (Figures 4a and 4b, encompassing 14 August 12 UTC to 16 August 00 UTC). At times, the smoke within the boundary layer almost fully extinguishes the lidar signal (e.g., 14 August 15 UCT), and the ceilometer-derived...
Figure 5. The 27-member ensemble HYSPLIT back trajectories initialized on 13 August 2016 12 UTC originating from Ascension Island at (a) 500 m, (b) 1 km, (c) 2 km, and (d) 3 km, driven by 0.5° NCEP GFS meteorology. The August 2016-mean ERA-Interim geopotential heights and vector winds are overlaid at (a) 1,000, (b) 900, (c) 800, and (d) 700 hPa [dashed black contours, and as closed color contours on panels (a)–(c)]. Panel (d) indicates the 3-km wind speeds in closed color contours. (e) One 500-m backtrajectory per day of August (12 UTC). (f) Spatial distribution of MODIS-detected fires for August 2016.
cloud base heights are included to distinguish aerosol from cloud. The pronounced extinctions coincide with relative humidity maxima, one at approximately 800 m, corresponding to the lifting condensation level, and 1.5 km just below the trade wind inversion (Figure 4c). These relative humidity profiles indicate a decoupled boundary layer, often observed at Ascension. Both cloud-containing layers are individually well mixed in moisture (not shown, but consistent with the linear increase in the relative humidity with altitude). The deliquescence of the smoke particles is confirmed by the lower lidar volume depolarization ratios within the boundary layer (Figure 4b) indicative of more spherical particles.

The near-surface water vapor mixing ratio of 12–14 g/kg decreases to ~3 g/kg above the trade wind inversion (not shown) for a relative humidity of 20-30%. The specific humidity of the upper aerosol layer indicates air that was last saturated at an altitude of approximately 5 km, consistent with the residual of a deep continental boundary layer. The lidar volume depolarization ratio increases, consistent with more desiccated, aspherical aerosol. The higher aerosol layer is resting directly upon the trade wind inversion, most obvious in Figure 4c; this feature was common to almost all the days with an upper level smoke layer (visually determined from daily lidar imagery). The lidar extinction values above the cloud layer are considered biased low by a factor of 0.35 (see supporting information) and the vertical gradient in extinction reflects attenuation of the lidar signal, as the moisture layer is well mixed. Either by assuming the extinction just above the cloud layer of 0.1 km$^{-1}$ is a constant through the aerosol layer, or by correcting by a factor of 2.8 supports an estimate for the above cloud aerosol optical depth of approximately 0.2 until midday 15 August, or almost one half of the column sun-photometer-derived aerosol optical depth of 0.48 between 15 and 18 UTC on 14 August.

Radiosonde-derived wind profiles indicate westward winds throughout the entire 0–4 km column during 13–16 August 2016 (not shown). The 7-day HYSPLIT backtrajectories at 500, 1,000, and 2,000 m indicate a direct northwestern transport from continental African fire source regions, with the week-long meteorology integrating to bring continental smoke to Ascension Island (Figure 5). This atmospheric boundary layer flow contrasts with the climatological wind pattern of southeasterlies advecting clean Southern Hemisphere air around the southern Atlantic subtropical anticyclone, apparent on backtrajectories for most other August days (Figure 5e). The 31 August is another day with both high near-surface rBC values and a 500-m backtrajectory tracing back to continental Africa. This flow pattern, previously noted in Swap et al. (1996), appears to be important for explaining high near-surface aerosol loadings despite continental air, originating from highlands with higher isentropic values, typically riding over the marine boundary layer.

5. Summary

Key points are

1. Near-surface rBC mass concentrations vary significantly at synoptic time scales from June to October at the Ascension Island location. These and their accompanying nonrefractory aerosol strongly affect the boundary layer radiative and microphysical environment.
2. A spectrally-flat absorption angstrom exponent suggests a lens coating of the black carbon is primarily responsible for the strong absorption, with the presence of additional aerosols modulating the light absorption seasonally. The most light absorption per rBC mass occurs in June, and the SSA increases from an August mean value of 0.78 ± 0.02 to 0.81 ± 0.03 in September to 0.83 ± 0.03 in October, at the green wavelength averaged over both 2016 and 2017.
3. The aerosol loadings within and above the cloudy boundary layer do not necessarily correlate well, with more of the total column aerosol present in the boundary layer early in the BBA season, migrating to predominantly free-tropospheric aerosol in September.
4. A campaign-maximum in near-surface rBC and total column aerosol optical depth on 13–16 August 2016 can be explained by more direct transport at lower altitudes of smoke off of the African continent.
5. Micropulse lidar observations indicate that upper level aerosol typically extends down to the trade wind inversion.

These early findings support an inference of a June maximum over the southeast Atlantic in a satellite-derived cloud droplet number concentration climatology (Bennartz & Rausch, 2017). The observation that more of the aerosol is above the cloudy boundary layer in September–October is more in keeping with the climatological monthly-mean MODIS clear-sky fine-mode aerosol optical depth distribution (Adebiyi et al., 2015). The findings also point to the need for continuing research. The predominant pathways by which smoke enters
the Ascension Island boundary layer for each month still remain to be established. The details of the aerosol composition and size affecting the hygroscopic and optical properties as they evolve over time, reflecting differences in source region, aging, and background conditions need further scrutiny. The vertical structure in the aerosol optical properties needs to be more robustly established and will benefit from measurements made by the UK CLARIFY (Clouds and Aerosol Radiative Impacts and Forcing: Year 2017) aircraft campaign deployed on Ascension in August-September, 2017. The dominant aerosol-cloud interactions still remain to be established and discriminated from meteorology.

Acknowledgments
P. Z., R. D. and J. Z. gratefully acknowledge support from the Department of Energy (DOE) Atmospheric System Research (ASR) LASIC planning grant DE-SC0013720. Support is acknowledged from the DOE Office of Biological and Environmental Sciences (OBER) under contract DE-SC0017204 to AJS through the ASR program and to SS and ACK through the ASR and ARM programs. Connor Flynn was supported as Aerosol Lifecycle Working Group Translator by the OBER of the U.S. DOE as part of the ARM Climate Research Facility and Office of Science Scientific User Facility. Paytsar Muradyan is supported by the ARM Climate Research Facility. We thank Brent Holben and the AERONET ARM Climate Research Facility. We thank Adeyemi Adebiyi for Figure 5f. We thank two anonymous reviewers for their thoughtful commentary. Acknowledgments to the individual data sets are provided in the Supplementary Information.

References
Bennartz, R., & Rausch, J. (2017). Global and regional estimates of warm cloud droplet number concentration based on 13 years of AQUA-MODIS observations. Atmospheric Chemistry and Physics, 17, 9815–9836. https://doi.org/10.5194/acp-17-9815-2017


