The Convolution of Dynamics and Moisture with the Presence of Shortwave Absorbing Aerosols over the Southeast Atlantic

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ABSTRACT

Biomass burning aerosols seasonally overlie the subtropical southeast Atlantic stratocumulus deck. Previous modeling and observational studies have postulated a semidirect effect whereby shortwave absorption by the aerosol warms and stabilizes the lower troposphere, thickening the low-level clouds. The focus herein is on the dynamical and moisture effects that may be convoluted with the semidirect effect. Almost-daily radiosonde data from remote St. Helena Island (15.9°S, 5.6°W), covering September–October 2000–11, are combined with daily spatial averages (encompassing the island) of the MODIS clear-sky fine-mode aerosol optical depth ($\tau_{\text{af}}$). Increases in $\tau_{\text{af}}$ are associated with increases in 750–500-hPa moisture content. The net maximum longwave cooling by moisture of almost 0.45 K day$^{-1}$ reduces the aerosol layer warming from shortwave absorption. ERA-Interim spatial composites show that polluted conditions are associated with a strengthening of a deep land-based anticyclone over southern Africa, facilitating the westward offshore transport of both smoke and moisture at 600 hPa. The shallower surface-based South Atlantic anticyclone exhibits a less pronounced shift to the northeast, strengthening the low-level coastal jet exiting into the stratocumulus deck and cooling 1000-hPa potential temperatures. Warm continental outflow further increases the 800-hPa potential temperatures ($\theta_{\text{800}}$), reinforcing the lower tropospheric stability ($\theta_{\text{800}} - \theta_{\text{1000}}$) over the stratocumulus deck. Enhanced southerly dry air advection also strengthens the cloud-top humidity inversion. The increased stability helps explain an observed decrease in cloud-top heights despite an anomalous reduction in subsidence. The changes to the horizontal dynamics enhance low-level cloudiness. These are separate but not necessarily distinct from an aerosol semidirect effect, encouraging care in attribution studies.

1. Introduction

Episodically during July through October, emissions from biomass burning (BB) in the southwestern African savannah are advected westward over the Atlantic Ocean. Most of the BB aerosols lie between 750 and 500 hPa, distinctively separated from, and overlying, one of the three major planetary stratocumulus decks in the world (Fig. 1). Shortwave absorption by the dark-colored aerosols warms and stabilizes the lower troposphere. A large-eddy simulation relates the stabilization immediately above cloud top to a thicker cloud deck (Johnson et al. 2004), a so-called semidirect aerosol effect (Hansen et al. 1997). Satellite observations indeed associate more overlying smoke with warmer (by about 1 K) 700-hPa temperatures and higher cloud liquid water paths (Wilcox 2010), supporting the inference of the semidirect effect. Both the simulation and observational studies also find lower cloud tops.

The modeling study of Johnson et al. (2004) did not examine the impact of aerosol located as far above the cloud deck as is observed for the southeast Atlantic, while observations consistent with the semidirect effect do not necessarily imply attribution. In addition, observational studies typically span the July to October months to enhance their sampling (e.g., Chand et al. 2009; Wilcox 2010), as also done here for Fig. 1. While arguably necessary, this may confound seasonal cycle with aerosol effects. At synoptic time scales, circulation
patterns that encourage offshore smoke transport may also impact the stratocumulus deck through changes to the large-scale thermodynamic environment that are independent of the presence of smoke. Another difficulty that is peculiar to observational studies focusing on free-tropospheric absorbing aerosols is that the lower-tropospheric stability can no longer serve as an explicit control variable for meteorology (e.g., Loeb and Schuster 2008), although arguably the sea surface temperature still can (e.g., Wilcox 2012).

Another observational approach for characterizing the semidirect effect combines passive emission tracers with satellite data for the same offshore transport pathway for a range of aerosol loadings (e.g., Brioude et al. 2009). This can be effective for fire emissions, since the number and intensity of fires can vary spatially and on short time scales. A prior identification of the prevailing circulation patterns affecting BB aerosol transport, and how such patterns influence the stratocumulus deck independently of the aerosol, encourages intuition for such analyses. Such an analysis is the topic of this paper, building on a similar study of the southeast Pacific (Painemal and Zuidema 2010).

A highlight of this study is its incorporation of radiosondes from St. Helena, a small island located approximately 1800 km offshore of continental Africa (15.9°S, 5.6°W; also indicated in Fig. 1), to the west of the main stratocumulus deck identified within Klein and Hartmann (1993). Radiosondes have been archived at their original resolution since July 1999. The island experiences monthly-mean MODIS low cloud covers between July and October exceeding 60% (Fig. 1a) and is also often subject to an elevated smoke layer (Fig. 1b). The radiosondes, when combined with satellite observations on aerosol, provide a unique observational dataset of the thermodynamic profiles associated with polluted and pristine conditions. The radiosonde dataset is large enough that most of the analysis can focus on September and October only, months whose selection is justified in section 3. The assessment of reanalysis datasets using radiosonde datasets further allows us to select the most appropriate reanalysis for characterizing the large-scale circulation.

The radiosondes also encourage an evaluation of any radiative impacts from the moisture characteristics of smoke plumes. These have not yet been explicitly considered in previous literatures. The aerosol layers elevated above the southeastern Atlantic have typically been depicted as dry in direct observational analysis (e.g., Wilcox 2010), numerical weather models (e.g., Randles and Ramaswamy 2010), and large-eddy simulations (Johnson et al. 2004), perhaps in analogy to the Saharan dust layers (e.g., Wong et al. 2009). However, observations from the South African Regional Science Initiative (SAFARI)-2000 document aged smoke that is also swollen through humidification (Magi and Hobbs 2003). Reanalyzed in situ data from coastal flights near northern Namibia during the UK-SAFARI 2000 campaign (Haywood et al. 2003b) also clearly demonstrate a correlation between aerosol and specific humidity (Fig. 2). These show that the aerosol sampled within BB plumes are associated with specific humidities ($q_v$).
greater than 2 g kg\(^{-1}\), while outside the smoke plumes the \(q_v\) values are less than 1 g kg\(^{-1}\). If present, anomalous moisture that is collocated with the smoke will have additional radiative consequences that should be considered, both in the shortwave and longwave.

The datasets, the radiative transfer model, and the methodology used are discussed in section 2. The July–October trends are highlighted in section 3, and section 4 discusses the compositing of all the collocated soundings into pristine, intermediate, and polluted samples. The radiative effects of midtropospheric moisture at St. Helena are assessed in section 5. Section 6 discusses the large-scale circulation pattern governing pristine and polluted time periods, and the implications for the cloud structure, while section 7 provides a summary.

2. Data, radiative transfer model, and methodology

a. Radiosondes

The UK Met Office launches Vaisala RS-80 radiosondes at St. Helena through an intergovernmental agreement on weekdays (excluding public holidays) at approximately 1100 UTC, from a station at an elevation of 435 m above sea level. The radiosondes measure temperature, relative humidity, and wind variables every 2 s, for a vertical resolution of \(\sim 0.5\) hPa. This study uses soundings from September and October of 2000 through 2011 (excluding 2001 because of lack of data) with each profile containing complete temperature, moisture, and wind measurements up to 400 hPa. The total of 457 soundings are thereafter regressed to 2-hPa vertical resolution.

b. MODIS aerosol data

The Terra Moderate Resolution Imaging Spectroradiometer (MODIS) sensor provides clear-sky retrievals of aerosol optical depth (\(\tau_a\)) at 550 nm (Kaufman et al. 1997; Tanré et al. 1997). The \(\tau_a\) at 550 nm is multiplied by the fine-mode aerosol fraction (Remer et al. 2005) to create a fine-mode aerosol optical depth (\(\tau_{af}\)), hereafter used as a proxy for smoke aerosol optical depth (e.g., Sakaeda et al. 2011). We use Collection 5 of the MOD08 aerosol products, included in the Level-3 MODIS atmosphere daily product (Hubanks et al. 2008), at a spatial resolution of \(1^\circ \times 1^\circ\) and available since 2000. The operational MODIS retrieval globally assumes a value of the single-scattering albedo, set to 0.9 for smoke within Collection 5 (Ichoku et al. 2003).

MODIS \(\tau_a\) values at 380 nm compare reasonably well (\(r = 0.84\)) with \(\tau_a\) derived from Aerosol Robotics Network (AERONET) sun photometers at Ascension Island (8°S, 14.5°W), located to the northwest of St. Helena (Satheesh et al. 2009). The primary advantage of MODIS is the broad-swath width (\(\sim 2300\) km) that increases the opportunities for clear-sky \(\tau_{af}\) retrievals within the often-cloudy environment surrounding St. Helena. Although numerous studies highlight issues with MODIS aerosol retrievals in the presence of (noncollocated) clouds (e.g., Wen et al. 2007; Loeb and Schuster 2008), for the purpose of this study we are more interested in distinguishing as many cases of polluted versus pristine conditions as possible, rather than focusing on detailed accuracy. The ability of MODIS to survey larger expanses daily is preferred to the much more spatially limited space-based lidar sampling, despite lidar’s ability to detect aerosol above cloud.

The September–October average aerosol optical depths from different domain sizes are assessed for different spatial regions to determine the optimal trade-off between sample size and representativeness (Fig. 3). The \(\tau_{af}\) values were further composited into terciles, defined as \(\tau_{af} \leq 0.1\) (pristine), \(0.1 < \tau_{af} \leq 0.2\) (intermediate), and \(\tau_{af} > 0.2\) (polluted). Locations to the east of St. Helena Island typically have increased aerosol loadings. Retrievals of \(\tau_{af}\) were available for 260 of the 793 calendar days within the 1° box encompassing St. Helena. A larger box containing St. Helena in its southwest corner (7.5°–17.5°S, 7.5°W–2.5°E, outlined in red) increases the available \(\tau_{af}\) sample size to 784 days. The \(\tau_{af}\) values averaged over the larger domain are

![Fig. 2. Nephelometer aerosol scattering vs specific humidity inside and outside biomass burning aerosol plumes, defined respectively, as 60-s averages in which \(\sigma_{\text{sca}} < 0.025\) m km\(^{-2}\).](image-url)
somewhat larger than for those centered over the island (0.35 vs 0.27 for the most polluted tercile).

Farther east, the $\tau_{af}$ averaged over the main stratocumulus region ($10^\circ$–$20^\circ$S, $10^\circ$W–$0^\circ$W; outlined in black in Fig. 3) identified by Klein and Hartmann (1993) correlates somewhat with those $\tau_{af}$ retrieved over St. Helena ($r = 0.45$), improving for those averaged over the larger domain ($r = 0.67$). The $\tau_{af}$ distributions for the three regions are skewed unimodal distributions (not shown) that vary in a systematic manner, and lack outliers. This suggests that results based on the larger middle domain encompassing St. Helena can also be relevant for the main stratocumulus region. We base our composite analysis using the larger domain containing St. Helena, primarily to increase the sample size (108, 253, and 423, for the three aerosol terciles). When matched with the radiosondes, the respective matched $\tau_{af}$–radiosonde pairs had sample sizes of 58, 159, and 232 (449 of the available 457 soundings).

c. CALIOP aerosol data

The primary disadvantage of the MODIS operational retrievals is that aerosols above clouds are not identified and no information on the aerosol vertical structure is provided. Space-based lidar data provide the relationship of the vertical structure of the elevated smoke layers to that of the radiosondes for those few but important days when the lidar sampled locations near St. Helena. The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) on board the Cloud Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO; launched on 28 April 2006) provides vertically resolved retrievals of volume extinction coefficients at 532 nm (Winker et al. 2003, 2009). The July–October 2006–12 vertically resolved smoke counts, shown in Fig. 1b, are derived from level 3 aerosol type histograms provided in 2° (latitude) × 5° (longitude) horizontal resolution, and 60-m vertical resolution (Omar et al. 2009). These show that in the mean, smoke occurs over St. Helena between ~750 and 480 hPa, peaking at ~620 hPa.

Examples of individual profiles of aerosol extinction drawn from each of the three aerosol groups show significant similarities with the sounding profiles of temperature and moisture, and interesting boundary layer features (Fig. 4). The example from 16 August 2006, from the most polluted tercile, has a pronounced smoke layer with the highest extinction occurring near the layer top, where it coincides with a strong temperature/moisture inversion. The cloudy boundary layer is well mixed. The second example, from 15 October 2008, is of a smoke layer that contains enough moisture to support cloud at the smoke-layer top. Cloud-containing aerosol layers are slightly unusual but not rare either. The boundary layer possesses two distinct moisture layers of relatively constant potential temperatures, suggesting that the radiatively driven turbulence from the cloud top is not reaching the surface-based mixed layer. The last example, from 5 August 2008, is from the most pristine tercile. The sounding shows two separate mixed layers in the boundary layer, with the surface mixed layer reaching 1 km in depth. The CALIOP imagery shows that the double-layered boundary layer is extensive. These observations indicate a richness to the poorly understood aerosol–cloud–radiation interactions for this region.

![FIG. 3. Spatial maps of the mean September–October $\tau_{af}$ distribution for different offshore aerosol conditions, composited into terciles based on the $\tau_{af}$ distribution of a 1° box encompassing St. Helena: (a) pristine ($\tau_{af} \leq 0.1$), (b) intermediate ($0.1 < \tau_{af} \leq 0.2$), and (c) polluted ($\tau_{af} > 0.2$). The star indicates the location of St. Helena Island with its mean $\tau_{af}$ shown at the top right part of each figure. The red box region is 7.5°–17.5°S, 7.5°W–2.5°E (encompassing St. Helena Island), and the black box is the main stratocumulus region identified by Klein and Hartmann (1993). Their respective domain-averaged $\tau_{af}$ are also given.](image-url)
d. Reanalysis data

Three daily reanalysis datasets were evaluated: the National Centers for Environmental Prediction–National Center for Atmospheric Research (NCEP–NCAR) reanalysis (Kalnay et al. 1996), the NASA Modern-Era Retrospective Analysis for Research and Application (MERRA; Rienecker et al. 2011), and the ERA-Interim reanalysis (Dee et al. 2011). The horizontal resolution of the NCEP, MERRA, and ERA-Interim datasets used

Fig. 4. (left) St. Helena sounding profiles with (middle) CALIOP extinction profiles averaged within a 2° box centered on the island. (right) CALIOP 532-nm total backscattered intensity imagery (scale included) with the approximate location of St. Helena indicated by the cross-hatched symbol. The three cases correspond to (a) 16 Aug 2006, $\tau_{df} = 0.621$, (b) 15 Oct 2008 $\tau_{df} = 0.18$, and (c) 5 Aug 2008, daytime $\tau_{df} = 0.071$. 
within this analysis is 2.5°, 1.25°, and 1.5°, respectively, with corresponding 17, 42, and 37 pressure levels.

An important question is whether or not the reanalyses assimilate aerosol-influenced information. All three reanalyses have the potential to assimilate the St. Helena radiosondes, if the radiosondes pass the internal data quality control measures specific to each reanalysis. Thus, if the St. Helena winds and temperatures are influenced by shortwave aerosol absorption, none of the reanalyses can be considered completely free of aerosol influences. Nevertheless, of the three reanalyses, the NCEP–NCAR reanalysis is the least sensitive to aerosol, by virtue of not assimilating radiances of any kind. The MERRA and ERA-Interim reanalyses do both assimilate radiances, using different data assimilation schemes. MERRA uses a three-dimensional variational assimilation that searches for the best fit between its model and selected sets of observations that include a full range of Earth Observing System satellite radiances. ERA-Interim uses a four-dimensional variational assimilation in which their model is projected both forward and backward until a best fit to a set of observations is achieved. Both MERRA and ERA-Interim directly assimilate aerosol-affected radiances from, for instance, the Atmospheric Infrared Sounder, meaning that their meteorological characterizations remain difficult to separate entirely from aerosol effects. In our analysis, we apply the convention that the reanalyses primarily reflect meteorology, but recognize we cannot completely exclude possible contributions from aerosol shortwave absorption through the reanalyses data assimilation procedures.

e. Radiative transfer model

Radiative transfer simulations investigated the relative radiative impact of moisture and absorbing aerosol for different underlying cloud optical thicknesses at St. Helena. These simulations were performed on each of the matched MODIS–radiosonde cases, using tercile-mean

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**FIG. 5.** (a) July, (b) August, (c) September, and (d) October MODIS mean 2002–12 cloud fraction (blue to black contours, 0.6–1.0 increments of 0.1), fine-mode aerosol optical depth (yellow-red shading indicates 0.25–0.45 in increments of 0.05 and very light black contour lines indicate 0.5–0.7 in increments of 0.1), fire pixel counts (green–red shading, 10–510 in increments of 50), and ERA-Interim 2002–12 monthly-mean 600-hPa winds. Red squares indicate Ascension Island and St. Helena Island.
thermodynamic conditions and aerosol optical depths that are vertically distributed according to the mean CALIOP-derived vertical structure. Since overlying absorbing aerosol introduce bias into operational MODIS derivations of cloud properties (Haywood et al. 2004; Meyer et al. 2013), the simulations used clouds with specified optical thicknesses $\tau_{\text{cloud}}$ of 5 and 15 instead, and a constant effective radius $r_e$ of 12 $\mu$m. The cloud volume extinction coefficients were distributed homogeneously between the mean CloudSat-derived cloud base at 0.5 km and cloud top at 1.2 km. The radiative impacts of fine vertical structure variations in moisture and aerosol, while likely important (e.g., Mapes and Zuidema 1996), were ignored here and left for future work.

The simulations utilized the Santa Barbara Discrete Ordinate Radiative Transfer (DISORT) Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al. 1998), chosen for its relative ease of use while containing the features needed for realistic computations. The shortwave and longwave bands span 0.3–1.02 $\mu$m and 4–20 $\mu$m, at spectral resolutions of 0.01 and 0.1 $\mu$m, respectively. The model incorporates the Discrete Ordinate Transfer model (Stamnes et al. 1988) and fluxes calculated here use the delta-four stream approximation (Wiscombe 1977). SBDART uses the Low Resolution Atmospheric Transmission Model (LOWTRAN 7; Kneizys et al. 1983; Pierluissi and Peng 1985) and does Mie scattering calculations for liquid cloud droplets and aerosols. The Lambertian ocean surface reflectance model is taken from Tanré et al. (1990).

SBDART includes four user-selected aerosol types, labeled oceanic, rural, urban, and troposphere (Shettle and Fenn 1979), but these cannot account for the wavelength dependence of BB aerosol radiative properties (Bond et al. 2013; Eck et al. 2003, 2013). These aerosol spectral properties depend on a complex internally mixed aerosol composition that includes, besides black carbon, primary and secondary organic aerosols. Eck et al. (2013) showed, using AERONET data, that the Ångström exponent for aerosols from biomass-burning regions of southern Africa has a range approximately between 1.7 and 2.0, with a mean of 1.85, over wavelengths of 440 to 870 nm. For this study, we use the mean Ångström exponent value (1.85) combined with the MODIS $\tau_{\text{af,550nm}}$ to compute $\tau_{\text{af}}$ values at 440 and 870 nm. These three wavelengths $\tau_{\text{af}}$ are specified for SBDART while logarithmic extrapolation is used to infer to other wavelengths across the visible range. The spectral asymmetry parameters of 0.66, 0.60, and 0.45 at 440, 550, and 870 nm, respectively, as determined during the SAFARI 2000 field campaign (Abel et al. 2005; Haywood et al. 2003b), are combined with Henyey Greenstein phase functions to determine the fraction of forward scattering and backscattering. These in situ derived asymmetry parameters are typically lower than the MODIS-retrieved values, which range from 0.75 to 0.78. Analysis (not shown) found this uncertainty to result in less than 10% change in the mean shortwave absorption values within the smoke layer.

For the single-scattering albedo ($\omega$), $\omega$ values are specified to be 0.86 at 550 nm, but allowed to vary by
±0.03 at that wavelength to estimate the impact of uncertainties in its specification. The σ values are extrapolated to the other shortwave wavelengths using a spectral dependence of −0.1 μm⁻¹, based on measurements from SAFARI 2000 (Eck et al. 2003) and following Wilcox (2010). The range of ±0.03 in the σ values at 550 nm reflect current uncertainties in known values, as single-scattering albedos cannot be remotely sensed from space. Haywood et al. (2003b), using primarily aircraft measurements near the northern Namibian coast, report σ_{550nm} values of smoke ranging between 0.84 and 0.91 with a mean of 0.9. Leahy et al. (2007) recommend that aircraft-derived σ_{550nm} measurements be revised down to 0.85 ± 0.02 based on comparisons to coastal AERONET stations. More recently, Eck et al. (2013) report a September mean σ_{550nm} of 0.87 using AERONET data from Mongu, Zambia. None of these studies explicitly considers aerosol aging. Abel et al. (2003) document σ_{550nm} values that increase from 0.84 at a source to about 0.9 in just 5 h (i.e., the smoke particles absorb less shortwave radiation as they age). Since the smoke over the Atlantic Ocean has been transported away from its source and can be considered “aged,” we use a σ_{550nm} value of 0.86 ± 0.03. This σ_{550nm} variability is also intended to capture a known seasonal trend in σ (Eck et al. 2013), thought to reflect changes in fuel load, as well as unaccounted biases (e.g., Bergstrom et al. 2007; Russell et al. 2010).

In summary, the radiative properties within SBDART are specified to reflect as closely as possible our best assessment of the literature on smoke radiative properties. No connection is explicitly made to the aerosol composition and environmental conditions. For example, while aerosol swelling is known to occur within the humid smoke plumes (Magi and Hobbs 2003), it is not explicitly accounted for here.
3. Subseasonal trends (July–October)

Trade-offs exist in determining the appropriate time span from which to articulate aerosol–cloud interactions. Longer time spans increase the sample size but they are more likely to contain meteorological variability and even subseasonal trends. Shorter time spans can select for specific meteorology, but may contain a statistically insignificant number of samples. In an initial analysis, we found that most pristine cases occurred in July, when the stratocumulus deck is small and most of the absorbing aerosols lie to the north of the cloud deck (Fig. 5). As the Southern Hemisphere moves from its winter into its spring, the continental fires shift southward and the

FIG. 8. ERA-Interim 600-hPa composite for (left) July–October and (right) the September–October minus July–August (SO-JA) difference for (a),(b) geopotential heights (m) and winds (m s\(^{-1}\)), (c),(d) temperature (K), (e),(f) specific humidity (g kg\(^{-1}\)), and (g),(h) pressure velocity (Pa min\(^{-1}\), positive values indicate subsidence). The star indicates the location of St. Helena and the main stratocumulus region identified by Klein and Hartmann (1993) is lightly contoured in black in (d),(f), and (h).
anticyclone over southern Africa strengthens. The combination establishes a September maximum in the continental aerosol outflow over the Atlantic basin, when the stratocumulus deck is also at its maximum size (Fig. 5). In October, both the aerosol outflow and the stratocumulus deck diminish in size as southern Africa moves into its summer. Thus most of the polluted cases occur in September. An analysis compositing on polluted/pristine cases drawing on the July–October time period may therefore reflect changes in the stratocumulus deck occurring between July (mostly pristine) and September (mostly polluted), which could be related to changes in large-scale circulation. Subseasonal changes are also apparent in the monthly-mean thermodynamic profiles for St. Helena, shown in Fig. 6 for July–October. A clear shift occurs from July–August to September–October, when the September–October boundary layer becomes shallower, and the water vapor content increases between 800 to 500 hPa.

Figure 7 depicts the Tropical Rainfall Measuring Mission (TRMM) rainfall climatology, highlighting the movement of the intertropical convergence zone.
southward in austral spring. Figures 8–10 show changes in the large-scale thermodynamic fields as the July–October mean and the difference of the September–October mean minus the July–August mean. As the region south of the equator moistens, the entire troposphere over central Africa cools, while south of 20°S the entire troposphere warms in response to the seasonal heating. The strengthening of the land-based anticyclone during September–October strengthens the 600-hPa easterlies at 10°S, and the free troposphere offshore also cools (Fig. 8). A large (3 g kg⁻¹) increase in the 600-hPa specific humidity overland at ~10°S reflects the increase in rainfall, with westward transport of the moisture apparent up to 10°W, encompassing St. Helena Island. At 800 hPa, the shallower South Atlantic anticyclone expands southward in the monthly progression, further allowing easterlies at 10°S to strengthen (Fig. 9) and inducing an anomalous anticyclonic circulation around St. Helena. Overall the 800-hPa surface warms by 2–3 K over almost the entire subtropical Atlantic basin, as
Fig. 11. St. Helena radiosonde (left) potential temperature ($\theta$), (middle) relative humidity (RH), and (right) specific humidity ($q_v$) individual (gray) and mean (black) profiles for (a)–(c) $\tau_{\text{af}} \leq 0.1$, (d)–(f) $0.1 < \tau_{\text{af}} \leq 0.2$, and (g)–(i) for $\tau_{\text{af}} > 0.2$, for September and October, 2000–11, with $N$ indicating the number of soundings contributing to each $\tau_{\text{af}}$ bin.
well as moistens. Near the surface, a southward shift in the South Atlantic high increases the southerly 1000-hPa winds flowing into the stratocumulus deck (Fig. 10) despite a slight weakening of sea level pressure center high. The near-surface air within the stratocumulus region cools, consistent with temperature advection from cooler latitudes. The entire Southern Hemisphere near-surface air moistens as it moves into its spring.

The monthly-mean radiosonde profiles (Fig. 6) show a clear increase in midtropospheric moisture in the same months that offshore aerosol transport to the west is maximized, suggesting an association between the aerosol and moisture. A substantial shift in the boundary layer is also evident between August to September, with the boundary layer depth shoaling and becoming moister and slightly cooler. Based on the radiosonde profiles and insights from the large-scale spatial climatologies, we chose to base the analysis on the September and October months only, with a 12-yr dataset still providing enough samples to support a statistical study.

4. Vertical structure at St. Helena

A visual examination of many aerosol extinction and humidity profiles revealed similarities in the vertical structure, especially in the broad 750–500-hPa layer where the aerosols and moisture coexist, at times with matching finescale vertical structures (e.g., Fig. 4). The aerosol layer is capped by the temperature–moisture inversions apparent in the coincident sounding profiles. Aircraft observations, taken closer to the African coast during UK SAFARI-2000, have shown similar features (Haywood et al. 2003a). A compositing of the radiosonde by aerosol optical depth quantifies the mean changes in the thermodynamic vertical structure. Average profiles of the potential temperature ($\theta$), specific humidity ($q_v$), and relative humidity (RH) for each $\tau_{af}$ tercile, using the daily-averaged $\tau_{af}$ of the domain 7.5°–17.5°S, 7.5°W–2.5°E, are shown in Fig. 11 along with the individual soundings.

All of the soundings show the pronounced low-level temperature and moisture inversions between 800 and 900 hPa that characterize subsiding stratocumulus regions. These inversions are shown to become stronger as the aerosol loadings increase. The boundary layers also become moister, by $\sim0.3$ g kg$^{-1}$ (Figs. 11ci), and slightly cooler. As a result, about 47% of the most polluted cases (109 out of 232) possess saturated boundary layers (RHs of 100%), compared to 31% of the most pristine profiles (18 out of 58).

Similarly, the midtroposphere is drier for the cases containing less aerosol, with a 750–500-hPa layer mean...
of 1.2 g kg$^{-1}$ for the pristine environments ($\tau_{af} \leq 0.1$; Figs. 11a–c), compared to 1.9 g kg$^{-1}$ for the most polluted case ($\tau_{af} > 0.2$). The few pristine environment profiles with $q_v > 1.2$ g kg$^{-1}$ occur on days preceding the inflow of pollution (not shown). For the more polluted soundings with $\tau_{af} > 0.2$ (Figs. 11g–i), the maximum $q_v$ values occur around 640 hPa in the mean, with some soundings placing the maximum near 500 hPa. Individual profiles with midtropospheric $q_v$ exceeding ~2 g kg$^{-1}$ occur more than 70% of the time during the most polluted conditions (Fig. 12). Relative humidities often exceed 60% (blue), usually close to the inversion capping the midtroposphere moisture. The relative humidity in several soundings is close to saturation, thus capable of activating aerosol into cloud condensation nuclei, explaining the occasional cloud feature observed in the midtroposphere (e.g., Fig. 4b).

The radiosondes provide our observational assessment of the NCEP-NCAR, MERRA, and ERA-Interim reanalyses. Averages over the terciles show that the reanalyses largely capture the mean thermodynamical structure of the soundings (Fig. 13), with the largest discrepancies between 700 and 1000 hPa, where the reanalyses with higher vertical resolutions (MERRA and ERA-Interim) perform better than NCEP, although all have difficulty representing the boundary layer-top inversions. Dynamically, the reanalyses all have stronger easterly winds than observed in the mid and lower troposphere, and weaker winds than observed directly above the boundary layer inversions.

Difference profiles, constructed by subtracting the pristine composite profile from the intermediate/polluted composite profiles, more cleanly highlight the differences in the vertical structure associated with the different $\tau_{af}$ regimes. The difference profiles are constructed as $\delta \psi = \langle \psi \rangle_{\tau_{af}} - \langle \psi \rangle_{\tau_{af} \leq 0.1}$, where $\psi$ is temperature and relative and specific humidity, and angle brackets imply the composite mean, shown in Figs. 14a–c for the soundings.
FIG. 14. (left) Temperature, (middle) relative humidity, and (right) specific humidity mean difference profiles calculated from the (a)–(c) UK radiosondes and (d)–(f) ERA-Interim, (g)–(i) NCEP, and (j)–(l) MERRA reanalysis data. The differences are constructed by subtracting the mean profiles corresponding to $\tau_{af} = 0.1$ from the other mean profiles shown in Fig. 11.
and Figs. 14d–l for the reanalyses. In the boundary layer, the most polluted radiosonde composite is the shallowest, coldest, and moistest of the three terciles shown (Figs. 14a–c). Both the humidity and temperature boundary layer-top inversions strengthen as the aerosol loading above increases. Wong et al. (2009) highlight similar boundary layer inversion features for the North Atlantic stratocumulus region, in association with elevated Saharan air layers. The warming at 700 hPa as aerosol loadings increase has also been previously noted (Wilcox 2010). Not previously documented, however, is that the specific humidity is also higher over a broad altitude range (750–500 hPa), for the most polluted composite. As $\tau_{af}$ increases, the moisture difference increases and broadens in height. Also not previously documented is the colder temperatures, by up to 2 K, at pressures, when biomass burning aerosols are present. The maximum cold temperature difference (Fig. 14a) resides near the top of the moisture anomaly (Fig. 14c) and contradicts the idea that shortwave absorption warms the full vertical extent of the aerosol layer. All of the features noted in the sounding differences are also evident in the large-scale composites (Figs. 8–10).

The corresponding radiosonde-matched ERA-Interim, NCEP–NCAR, and MERRA reanalysis difference composites (Figs. 14d–l) resemble the sounding differences. MERRA and ERA-Interim clearly match the magnitude of the observed vertical structure variations better than does NCEP, reflecting a more sophisticated but also more aerosol-influenced data assimilation approach. The ERA-Interim reanalysis reflects the ordering of the observed 800–1000-hPa mean specific humidity profiles as a function of the aerosol loading more accurately than does the MERRA reanalysis, and the ERA-Interim boundary layer temperatures do not contain the incorrect warming evident in the MERRA results. For these reasons, the ERA-Interim reanalysis is selected for all the depictions of the large-scale circulation presented here. Similar weaker depictions have also been produced using NCEP–NCAR reanalysis (not shown), supporting a meteorological interpretation.

The mean radiosonde and ERA-Interim zonal and meridional winds as a function of $\tau_{af}$ are shown in Fig. 15. In the midtroposphere, the observed winds weaken ($\sim 3 \text{ m s}^{-1}$) and shift from slight westerlies to northerlies as the aerosol loadings increase. Near the boundary layer top the observed southeasterly winds reach $\sim 10 \text{ m s}^{-1}$. The reanalysis boundary layer easterlies are slightly weaker here, and do not fully capture the wind shear. In the midtroposphere the ERA-Interim winds reflect the observed winds fairly well, although they are mildly easterly for polluted cases when the observed mean zonal winds are calm. The ERA-Interim reanalysis also includes a depiction of the nonobserved free-tropospheric subsidence: a reduction when more absorbing aerosols are present, also seen in the model results of Sakaeda et al. (2011). This would suggest a deeper boundary layer as a result, all else equal, but the radiosonde observations clearly depict a shoaling of
5. Radiative impact of the midtropospheric moisture

A midtroposphere that is more moist will experience an additional radiative heating in both the longwave (LW) and the shortwave (SW), independent of the presence of biomass burning aerosol. Three simulations are performed to investigate the radiative effect of the midtropospheric moisture. The three simulations include a clear sky and two cloudy skies, with cloud optical depths (\( \tau_{\text{cloud}} \)) of 5 and 15, both at a constant cloud effective radius (\( r_e \)) of 12 microns. Each experiment is done using two of the mean thermodynamic composite profiles shown in Fig. 11: a “dry” simulation corresponding to a pristine environment with little midtroposphere moisture (Figs. 11a–c) and a “moist” simulation corresponding to \( \tau_{\text{af}} > 0.2 \), incorporating a 700–500-hPa layer-mean increase in midtropospheric moisture of 0.83 g kg\(^{-1}\) (Figs. 11g–i). The background thermodynamical profiles establish the vertical resolution of 15.25 hPa used in the simulations. The aerosol volume extinction profile is assumed to vary vertically according to the mean CALIOP aerosol density profile. Radiative transfer computations are done for each member of the radiosonde–MODIS \( \tau_{\text{af}} \) collocated dataset and then averaged. The solar zenith angle is internally calculated using the time of the day and the St. Helena location. We compute daily averages of the instantaneous heating rate from 3-hourly calculations done between 0000 and 2100 UTC using the same thermodynamical profiles at each time, lacking other information.
In clear sky, the moisture absorbs shortwave radiation in addition to that by BB aerosol alone (Fig. 16). The maximum increase in shortwave heating due to moisture is approximately $0.12 \text{ K day}^{-1}$, compared to an aerosol SW heating rate of up to $1.2 \text{ K day}^{-1}$. Variations in $\sigma$ of $\pm 0.03$ affect the moisture layer-mean SW heating rate by $\pm 9\% - 23\%$, increasing with aerosol loading. In Fig. 17, the shortwave and longwave heating rates are shown, averaged over all of the aerosol optical depths and $\sigma$ of 0.86, for the three different cloud optical depths (0, 5, 15). When an underlying cloud is present, multiple reflection between the aerosol layer and the cloud layer increases opportunities for absorption by both the water vapor and aerosol and the shortwave heating increases further (Fig. 17). This increases as the cloud becomes more opaque, corresponding to a $\left[\frac{\delta(\text{aerosol heating})}{\delta(\tau_{\text{cloud}})}\right]$ of $\sim (0.2 \text{ K day}^{-1})/10$. This represents a positive feedback: if the shortwave absorption by the BB aerosol stabilizes the atmosphere immediately above the cloud, semidirectly thickening the cloud, the cloud in turn reflects more shortwave radiation back into the aerosol layer, further heating the aerosol layer.

Smoke aerosol is too fine to be radiatively active at infrared wavelengths, and only the increased midtropospheric moisture increases the longwave radiative cooling of the aerosol layer (Fig. 17b). This is only slightly reduced (by $\sim 10\%$), when underlying boundary layer clouds are introduced. The reduction occurs due the cooler cloud tops, which reduce the infrared absorption from below. Figure 18 summarizes the differences between the moist and dry simulations for the shortwave, longwave, and the net radiative heating, for both clear
and cloudy skies, averaged over all aerosol optical depths. The most notable feature is that the net radiative effect of the additional moisture is a cooling. The maximum of the net cooling, of almost 0.45 K day$^{-1}$, is located near the top of the moisture layer. This reduces the impact of the shortwave aerosol absorption, which reaches a maximum of 1.5 K day$^{-1}$, by approximately one-third.

Midtropospheric moisture in combination with aerosol not only radiatively affects the aerosol layer but also reduces the radiation reaching the boundary layer. Figure 19 summarizes the diurnal-mean instantaneous SW heating rate averaged over the smoke layer (736–469 hPa) and boundary layer (defined as 954–877 hPa, given that the station elevation at St. Helena is 435 m) as a function of aerosol optical depth. Boundary layers with thicker clouds, in which the cloud liquid water also absorbs shortwave radiation, are more sensitive to the attenuation. The increase in midtropospheric moisture from pristine to polluted conditions reduces the boundary layer SW absorption by 0.1 K day$^{-1}$. This more than doubles the reduction by 0.05 K day$^{-1}$ in boundary layer SW absorption from changes in $\tau_{af}$ from 0.1 to 0.3.

The increase in midtropospheric moisture also has an impact on the cloud-top longwave cooling. For an additional midtropospheric moisture of about 1.2 g kg$^{-1}$, the downwelling longwave radiation averaged between 550 and 750 hPa increases by about 15 W m$^{-2}$, reducing the net longwave cloud-top cooling by the same amount for a cloud of the same optical depth. The change in both the SW and LW radiative heating structure within the boundary layer from changes in midtropospheric moisture associated with the increased aerosol loading, will feedback further on cloud processes such as shortwave-induced decoupling and longwave-induced turbulence production.

6. Spatial patterns as a function of aerosol loading

Large-scale spatial patterns of the circulation, temperature, and moisture fields for pristine and polluted conditions at 1000, 800, and 600 hPa, corresponding to the surface, above cloud top and the upper BB aerosol layer, respectively (Fig. 14, and consistent with Figs. 8–10), are interpreted from a dynamical viewpoint. The composite of each aerosol bin is subtracted from the September–October mean: $\Delta \psi = \langle \psi \rangle_{\tau_{af}} - \langle \psi \rangle_{\text{Sept-Oct}}$, where $\psi$ is geopotential heights, winds, temperature, or specific humidity and the angle brackets $\langle \rangle$ indicate the composite mean (shown in Figs. 20, 21, and 23).
The 800-hPa temperature and moisture advection pattern (Fig. 22) is also shown to help with interpretation. These composites are then connected to observations of the all-sky albedo, cloud fraction, and surface winds.

At 600 hPa, changes in aerosol conditions can be associated with a strengthening of the 600-hPa anticyclonic pattern over the subtropical Atlantic (Fig. 20). The geopotential heights are higher and the prevailing easterlies stronger (Figs. 20a,b; see also Fig. 8), with colder temperatures and increased moisture over the main stratocumulus region and to its west (Figs. 20c–f). The temperature and specific humidity changes are consistent with moisture and anomalous cold temperature advection off of the continent (Fig. 8).

At 800 hPa (Fig. 21), the geopotential heights and easterlies are also stronger in the near-coastal region, during more polluted conditions, due to a westward shift of the continental-based anticyclone toward the offshore counterpart. The stratocumulus region warms and dries at 800 hPa as the aerosol loading increases, in contrast to the 600-hPa level. The temperature and moisture advection fields at 800 hPa are shown in Fig. 22, to help characterize the sources for the warming and drying. The strong north–south moisture gradient shifts north in response to a strong southerly flux of dry air over the main stratocumulus region (Figs. 22d,g). This southerly dry air advection provides an alternative explanation (from subsidence) of previously observed near-coastal clear-air slots separating the BB aerosol layer from the underlying clouds during SAFARI-2000 (Hobbs 2003). Horizontal temperature advection anomalously warms the 800-hPa level during polluted conditions, by about 0.2 K day$^{-1}$ averaged over the stratocumulus region, but the reduction in subsidence generates a net dynamical anomalous cooling over the same region (Figs. 22h,i) of about 0.5 K day$^{-1}$, despite an increase in the static stability (e.g., Fig. 14). The net anomalous cooling from the horizontal and vertical temperature advection is not consistent with the warmer 800-hPa temperatures shown in Fig. 21d for polluted conditions. A similar analysis at 700 hPa attributed a “missing heating” to aerosol shortwave absorption (Wilcox 2010); however, at 800 hPa direct radiative heating from aerosol absorption is much less than at 600 hPa, where more of the aerosol resides. It is in fact near zero within our radiative simulations based on the mean CALIOP aerosol density vertical distribution. If the cause is aerosol absorption, a mechanism would be needed to communicate the warming at 600 to 800 hPa.

Such a mechanism could explain the increase in the static stability at 800 hPa for more polluted conditions (Fig. 14), one that is underrepresented in the ERA-Interim reanalysis. Another possible explanation could be an underestimated horizontal temperature advection closer to the actual cloud top, consistent with reanalysis winds that are weaker than observed near cloud top (Fig. 15). For now, we merely note that the changes in the dynamical system that encourage reduced subsidence at 800 hPa also appear to facilitate the horizontal temperature advection over the same region (Figs. 21h,i), but that more careful attribution studies will be needed to relate the dynamical changes to the changes in the stability near cloud top.
Figure 23 shows the surface circulation patterns and thermodynamical changes associated with changes in aerosol conditions aloft. The South Atlantic anticyclone is shifted slightly to the northeast during more polluted conditions, encouraging a weak strengthening of the near-coastal winds south of 20°S. The stronger southerly winds imply more cold temperature advection near the surface, and can explain the spatial pattern of the cooler surface temperature south of 15°S in Fig. 23d. Overall, surface moisture increases, also consistent with increased surface fluxes south of 15°S induced by the stronger winds, although a more detailed budget is needed to attribute the moisture source north of 15°S.

Observations further clarify changes in spatial features with aerosol loading. The top-of-atmosphere all-sky albedo from the Clouds and the Earth’s Radiant Energy System (CERES) increases for more polluted conditions, despite the presence of the shortwave-absorbing aerosols (Fig. 24b), but consistent with
increased cloudiness below the aerosol layer. This is confirmed by a MODIS-derived cloud amount increase, which closely follows the contours of lower tropospheric stability (LTS; $\theta_{800\,\text{hPa}} - \theta_{1000\,\text{hPa}}$), calculated from ERA-Interim using the lower-altitude $\theta_{800\,\text{hPa}}$ to avoid the smoke-bearing layer. Although some of the cloud fraction increase may be spurious (aerosol interpreted as cloud), the increase must be qualitatively correct to explain the increase in CERES top-of-atmosphere all-sky albedo. The LTS increases by approximately 2 K over the main stratocumulus region, extending westward to about $-20^\circ$W during polluted conditions (see also Fig. 22), whereas under the more pristine case the cloud deck extent is limited to $-10^\circ$W. The enhanced low cloudiness typifying polluted conditions provides a positive feedback through the cloud-top radiative cooling to the strength of the cloud-top inversion, but the initial cause of a strengthened cloud-top inversion is not revealed within our analysis, and must be left to future work.

QuikSCAT-derived surface winds identify the coastal wind maximum as a low-level jet (see also Nicholson 2010). This is also present in the ERA-Interim reanalysis, but is not as well resolved (Fig. 23b). Changes in the observed QuikSCAT near-coastal winds are also supportive of the stratocumulus deck, with the low-level
jet along the Namibian coast increasing by up to 1.25 m s$^{-2}$ (Fig. 24f) in response to the enhanced zonal pressure gradient from the shifted South Atlantic anticyclone. Similar to the southeast Pacific (Xu et al. 2005), this should act to increase near-surface cold air advection into the southern edge of the stratocumulus region, intensifying surface sensible heat fluxes.

Previous studies discussing changes in cloud-top height ($z_{\text{cloud-top}}$) with aerosol loading are not in full agreement, with Johnson et al. (2004) and Wilcox (2010) reporting decreased cloud top heights in the presence of absorbing aerosol above the cloud top, consistent with enhanced stability and reduced cloud-top entrainment. Sakaeda et al. (2011), however, report a cloud-top height increase in their modeling study, attributed to reduced free-tropospheric subsidence. We evaluate changes in cloud-top height with aerosol loading using both the soundings at St. Helena and a satellite-derived measure. An inversion-base height is inferred from a radiosonde RH $\geq$ 95% and a $d(RH)/dP \geq 9\%/4$ hPa below $\sim$750 hPa, serving as a proxy for cloud-top height. The relative frequency of the inversion-base heights, composited by MODIS $\tau_{ul}$, shows that the mean inversion base is lower by about 93 m in polluted conditions (Fig. 25) and is also more sharply defined in the vertical. We also estimate the $z_{\text{cloud-top}}$ for the southeast Atlantic following the relationship derived within Zuidema et al. (2009) using MODIS cloud-top temperature and TRMM Microwave Imager sea surface temperature (Figs. 25b–d). Only pixels with cloud-top temperatures greater than 0.8°C and cloud fractions greater than 0.9 are assessed. These show a decrease of about 112 m in $z_{\text{cloud-top}}$ near St. Helena under more polluted conditions. The maximum decrease in $z_{\text{cloud-top}}$ occurs south of 15°S, possibly as a dynamical response to the northward displaced coastal jet (e.g., Muñoz and Garreaud 2005). The lowered cloud-top heights are consistent with the observations of Wilcox (2010), but a good attribution to changes...
in the large-scale temperature advection or to the aerosol radiative heating remains lacking (Figs. 9 and 21).

7. Summary

The following summarizes the results found in this study:

- A clear shift in the thermodynamical structure and circulation patterns is apparent between July–August and September–October composites. As the Southern Hemisphere warms, tropical precipitation moves southward, and the midtropospheric anticyclone over southern Africa strengthens. September is when absorbing aerosols overlying stratocumulus are most prevalent.
- ERA-Interim reanalysis thermodynamic profiles compared better to radiosondes composited by daily MODIS fine-mode aerosol optical depth ($\tau_{a}$) than do MERRA and NCEP–NCAR profiles, at St. Helena Island (15.9°S, 5.6°W).
- Moisture is also present within the elevated biomass burning (BB) aerosol layer (~750–500 hPa). The finescale vertical structure of individual radiosonde moisture profiles is often similar to that of CALIOP aerosol extinction profiles from near St. Helena. The moisture/smoke layers are typically capped by a sharply defined temperature–moisture inversion.
The midtropospheric moisture and $\tau_{af}$ increases can be linked to stronger anticyclones over the southern Africa and southern Atlantic regions. These drive enhanced easterly winds, localized at $\sim 10^5S$, during more polluted conditions that appear to transport BB aerosol and moisture far offshore.

The radiosonde composite from polluted days reveals a previously documented warmer temperature anomaly between 850 and 700 hPa, capped by a colder anomaly between 700 and 500 hPa. The cold anomaly has not been previously documented, and it contradicts the idea that shortwave absorption warms the full vertical extent of the biomass burning aerosol layer.

Radiative transfer calculations based on the radiosonde composites from St. Helena combined with the full range of retrieved $\tau_{af}$ indicate that the presence of midtropospheric moisture produces a net diurnal-mean anomalous cooling, reducing the impact of shortwave warming from BB aerosol alone from a maximum of $\sim 1.5$ to $\sim 1K day^{-1}$ at the peak of the

Fig. 24. 2000–11 September–October mean (a),(b) CERES all-sky albedo > 0.25, (c),(d) MODIS cloud fraction > 0.5, ERA lower-tropospheric static stability [LTS (K); green contour; defined as $\theta_{850} - \theta_{1000} > 14 K$; and (e),(f) QuikSCAT surface wind speed (>7 m s$^{-1}$; 2000–09) during (left) pristine ($\tau_{af} < 0.1$) and (right) polluted conditions ($\tau_{af} > 0.2$). The red box encompasses $7.5^5–17.5^5S$, 7.5°W–2.5°E, and the black box the main stratocumulus deck. Their respective average albedo and wind speed values are indicated in (a),(b) and (e),(f). The average MODIS cloud fractions within the red boxed area for pristine and polluted conditions are 0.79 and 0.85, and 0.82 and 0.86 for the black boxed area.
heating rate profile, averaged over all $\tau_{af}$ for the range of simulations considered.

- Warmer temperatures directly above the stratocumulus deck at 800 hPa, consistent with the westward advection of warmer temperatures from the continent, combined with cooler temperatures at the surface south of 15°S, act to increase the LTS ($\theta_{800} - \theta_{1000}$) by about 2 K. The wind speeds within a low-level coastal jet feeding into the stratocumulus region (south of 15°S) are also increased by up to 1.25 m s$^{-1}$. The enhanced low-level jet is consistent with a northeastward shift of the subtropical high, increasing the near-coast zonal pressure gradient.

- Drying at 800 hPa can be explained as a southerly advection of drier air, providing an alternative explanation (from subsidence) for previously observed clear-air slots separating the cloud layer from the smoky layer aloft (Hobbs 2003).

- The top-of-atmosphere albedo is increased when the absorbing aerosols are present, consistent with an increase in MODIS-inferred cloud fraction, and the observed stratocumulus clouds tops are clearly lower, as indicated in soundings and satellite retrievals. This decrease in cloud-top heights occurs despite reduced ERA-Interim subsidence but is consistent with the large-scale horizontal temperature advection.

- The increase in 800-hPa temperatures under polluted conditions is not fully explained by our dynamical analysis, which finds a net anomalous cooling due to the reduction in subsidence. Further work is required to distinguish the mechanism by which radiative heating aloft enhances the static stability near the cloud top, separate from the dynamical heating at the same level.

Nothing shown here discounts the semidirect effect, and indeed meteorological effects cannot be fully separated from the aerosol effects using observational data alone. Changes in dynamics and thermodynamics documented within this study mostly act in concert with the expectation of the aerosol semidirect effect, except for the reduction in subsidence, but contribute alternative explanations for changes in cloud fraction and depth. The net radiative cooling by the midtropospheric moisture partially compensates for any stabilization of the free troposphere caused by the aerosol shortwave absorption. We note that our study does not explicitly take into account the feedback processes that may arise from change in cloud or surface properties as a result of aerosol loading (e.g., Cook and Highwood 2004), or the impact of aerosol hygroscopicity in the vicinity of high relative humidity (e.g., Magi and Hobbs 2003). More in-depth study, facilitated by constrained modeling with and without absorbing aerosols present, along with a full

**FIG. 25.** (a) Relative frequency of St. Helena sonde-derived inversion base as a function of collocated MODIS $\tau_{af}$. (b) September–October mean satellite-derived cloud-top height for all $\tau_{af}$. Thick black contours indicate mean sea surface temperature values from the TRMM Microwave Imager. (c) Mean cloud-top height difference between polluted ($\tau_{af} > 0.2$) and pristine ($\tau_{af} \leq 0.1$) conditions.
budget and Lagrangian analyses, will be needed to more completely attribute cloudiness changes as either adjustments to the presence of aerosol aloft, or to the moisture aloft, or to associated changes in dynamics. Such efforts will benefit from a comprehensive and detailed characterization of the cloudy boundary layer and the aerosol spectral radiative properties over the remote southeast Atlantic.

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