Examining absorbing aerosols above clouds based on in situ and satellite observations

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EXAMINING ABSORBING AEROSOLS ABOVE CLOUDS

BASED ON IN SITU AND SATELLITE OBSERVATIONS

by

IAN Y. CHANG

A DISSERTATION

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for the degree of Doctor of Philosophy
in
The Department of Atmospheric Science
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The University of Alabama in Huntsville

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ABSTRACT
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Name of Candidate __________ Ian Y. Chang ________________________________
Title __ Examining absorbing aerosols above clouds based on in situ and satellite observations

The representation of aerosols, clouds, and aerosol-cloud radiative effects remains highly uncertain, restricting the reconstruction of past climate and the prediction of future climate change. The above-cloud absorbing aerosol direct radiative effects generally result in a positive sign at top-of-atmosphere (TOA) over the ocean. This sign results from a reduction of scattered radiation at TOA due to absorbing aerosol's SW absorption. Geostationary satellite data from the Spinning Enhanced Visible and Infrared Imager (SEVIRI) in conjunction with NASA A-Train data are used to develop an algorithm for detecting biomass burning smoke aerosols above clouds. The detection relies on spectral signatures, textural characteristics, and time-dependent spectral variation of SEVIRI data. The user accuracy of this algorithm is ~65% and the producer accuracy is over ~77%. Above-cloud AOD retrievals from SEVIRI and Moderate Resolution Imaging Spectroradiometer (MODIS) are compared against the Spectrometers for Sky-Scanning, Sun-Tracking Atmospheric Research (4STAR) data set for September 2016. MODIS and SEVIRI AODs have a good agreement compared with 4STAR AODs when simultaneously retrieved above-cloud AODs and underlying cloud properties assuming a single scattering albedo (SSA) of 0.84 at 470 nm and 0.76 at 860 nm. Above-cloud AOD retrieval generally agrees to within 0.2 above homogeneous clouds with a cloud optical depth (COD) greater than 8. However, above-cloud AOD retrievals are biased high when retrieving above thin clouds since the radiance separation between aerosols and clouds becomes complex. Results indicate that the assumed SSA play a significant role in the retrieved above-cloud AODs.
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CHAPTER ONE

INTRODUCTION

1.1 Overview

The Earth’s climate is affected by continuous changes of various atmospheric agents such
as greenhouse gases, aerosols, and albedo. The total well-mixed greenhouse gas (e.g., CO$_2$, CH$_4$,
N$_2$O, and halocarbons) forcing is estimated to be $+2.83$ Wm$^{-2}$ according to the Fifth Assessment
Report (AR5) of the Intergovernmental Panel on Climate Change (IPCC, 2013), which is an
increase from $+2.63$ Wm$^{-2}$ since the Fourth Assessment Report (AR4) (IPCC, 2007) due to
increased greenhouse gas concentrations. However, water vapor serves as the strongest
greenhouse effect on Earth’s atmosphere and its concentration depends strongly on temperature.
The amount of water vapor in saturated air increases as the temperature increases. Thus, sources
that alter temperature subsequently modify water vapor concentration, creating a positive
feedback (Hartmann, 1994; Held & Soden, 2000; IPCC, 2013).

The radiative effect by aerosols due to scattering and absorption of shortwave radiation is
known as the aerosol direct radiative effect (Loeb & Kato, 2002; Satheesh et al., 1999). When
radiative perturbation accounts for anthropogenic components between the aerosols of present
day and the pre-industrial era (i.e., 1750), it is known as the aerosol direct radiative forcing (Yu
et al., 2006; Loeb & Su, 2010). The aerosol direct radiative forcing is synonymous to “aerosol-
radiation interactions” (IPCC, 2013) and is estimated to have a negative forcing of –0.35 Wm\(^{-2}\). AR5 introduced the term “aerosol effective radiative forcing,” which is the aerosol radiative forcing that accounts for both the semi-direct and the indirect radiative effects in the General Circulation Model (GCM) and the Chemical Transport Model (CTM) Results from these models suggest that the effective radiative forcing for both aerosol-radiation interactions and aerosol-cloud interactions is –0.45 Wm\(^{-2}\).

Aerosols can also modify cloud properties via “aerosol-cloud interactions.” For example, absorbing aerosols modify thermodynamic stability of the atmosphere (Jacobson, 2012), which affect cloud properties via the semi-direct forcing (Johnson et al., 2004; Wilcox, 2012). Aerosols can either alter cloud microphysical properties via the first indirect effect (Twomey, 1974, 1977) or cloud lifetime via the second indirect effect (Albrecht, 1989). The net effects among direct, semi-direct, and the indirect effects further complicate the estimate of the total aerosol radiative effect (Zhou et al., 2017). AR5 only evaluated the effective radiative forcing of the aerosol-cloud interactions is identical to the effective radiative forcing of aerosol-radiation interactions (–0.45 Wm\(^{-2}\)).

The representation of aerosols, clouds, and aerosol-cloud radiative effects remains highly uncertain, restricting the reconstruction of past climate and the prediction of future climate change (Kahn, 2012; Myhre et al., 2013). Aerosols partially offset the greenhouse gas forcing but dominate the uncertainties of anthropogenic radiative forcing. For instance, aerosol-radiation interactions has an uncertainty ranging from –0.95 W m\(^{-2}\) to +0.05 W m\(^{-2}\) and aerosol-cloud interactions has an uncertainty ranging from –1.2 W m\(^{-2}\) to 0.0 W m\(^{-2}\).

Calculating the aerosol direct radiative effect requires an estimate of the aerosol optical depth (AOD) or synonymously as aerosol optical thickness (AOT), which is defined as the
aerosol extinction (scattering and absorption) in an integrated vertical column. Ratio of scattering to extinction (i.e., single scattering albedo) and relative directional scattering (phase functions) of aerosol particles are also important for quantifying the aerosol radiative effects (Yu et al., 2006). AODs can be estimated from in-situ field experiments, ground-based networks, satellite measurements, and numerical modeling experiments. For example, the aircraft in-situ instrument can infer AOD via direct solar beam transmittance below the aerosol layer (Shinozuka et al., 2013). Ground-based network such as the Aerosol Robotic Network (AERONET) provides capabilities for measuring AODs in cloud-free condition at high temporal frequencies (Holben et al., 1998). Satellite-derived AODs rely on the top-of-atmosphere (TOA) radiance inversion from look-up-tables (LUTs) (Gupta et al., 2016; Levy et al., 2013). Satellite sensors such as Multiangle Imaging SpectroRadiometer (MISR) provides multi-angle and multi-spectral measurements to infer aerosol properties (Diner et al., 1998; Kahn & Gaitley, 2015). The POLDER sensor provides polarized radiance to better infer particle size and shape (Tanré et al., 2011). Chemical transport models and global climate model simulates aerosol properties by accounting for aerosol emission, transportation, and deposition at various spatiotemporal scales (Myhre et al., 2013, 2017). Satellite observations can also be used to constrain model emissions for obtaining a more accurate emission source (Wang et al., 2016). The upcoming launch of Tropospheric emissions: Monitoring of Pollution (TEMPO) (Zoogman et al., 2017), a geostationary hyperspectral (from ultraviolet to visible) sensor, aims to monitor pollution in North America at an hourly basis, offering an unprecedented capability to study diurnal variations of pollution.

Current satellite operational aerosol products are limited to cloud-free conditions while above-cloud aerosol products are under development. Consolidating aerosols in cloud-free and
above-cloud conditions would yield a more realistic estimate of aerosol radiative effect. The mean aerosol direct radiative effect for absorbing aerosols above clouds is generally positive at the top-of-atmosphere (de Graaf et al., 2014; Wilcox, 2012), opposing the direct radiative effects in cloud-free regions. Methods for calculating the direct radiative effects of aerosols by accounting for aerosol-induced biases has been implemented (Meyer et al., 2013). Satellite observations over the southeast Atlantic have shown that changes in cloud fractions also produce biases in daily mean all-sky direct radiative effect (Min & Zhang, 2014). Aerosol heating modifies the lapse rate via the semi-direct effect (Johnson et al., 2004). Cooling due to negative semi-direct forcing opposed 60% of warming associated with the direct effect according to satellite data analyses (Wilcox, 2012). Aerosols can also act as cloud condensation nuclei and increase droplet number concentration and cloud albedo (Twomey, 1974) and alter both liquid water path and precipitation efficiency depending on the vertical separation distances between aerosols and clouds (Costantino & Bréon, 2013). These studies demonstrate the challenges and complexities in mapping and understanding the role that aerosols above or within clouds play in climate forcing.

Although all previous studies agree with the presence of positive radiative forcing for aerosols above clouds in southeast Atlantic, forcing magnitudes vary significantly on daily, seasonal, and interannual scales (de Graaf et al., 2014). Spatial distributions of above-cloud absorbing aerosols remain highly uncertain. Furthermore, because of the polar-orbiting nature, the temporal resolution of A-Train satellites is restricted to one overpass at a particular location per day during daytime, inhibiting above-cloud aerosol studies on a sub-daily scale. This study will carefully assess the radiative effects of above-cloud and within-cloud aerosols from a multifaceted approach by integrating various satellite sensors to leverage the spatiotemporal
analyses of this research area. Radiative transfer (RT) calculations will be applied to assess the radiative impact of aerosols above clouds and its heating rates, which can be used as a benchmark for modeling studies. The ObseRvations of Aerosols above CLouds and their intEracionS (ORACLES) field experiment offers an opportunity to assess uncertainties and improve satellite retrievals of aerosols and clouds and their radiative effects. Note that AOTs and AODs have the same definition and are used interchangeably in this study. Likewise, the terms cloud optical depths (CODs) and cloud optical thicknesses (COTs) are also used interchangeably in this study.

1.2. Outline

The overarching goal of this study is to expand our capabilities for studying aerosol-cloud radiative effects using satellite remote sensing, ground-based observations, radiative transfer models, and in situ measurements. First, this study examines the sensitivity of seasonalities on aerosols above clouds. The second goal is to develop an algorithm for detecting aerosols above clouds from the Spinning Enhanced Visible and Infrared Imager (SEVIRI). Finally, the goal is to validate the satellite-retrieved AODs against in-situ aircraft measurements. The specific objectives of this study is as follows:

1) Calculate the impact of seasonalities on above-cloud aerosol direct radiative effects and radiative heating rates from a delta-four stream RT model. Diurnal variations of radiative fluxes and radiative heating rates between June – October are examined using aerosol properties data from ground-based and in-situ observations. Satellite climatology of aerosol and cloud optical properties are also used to consolidate RT calculations.
2) Develop an algorithm for detecting absorbing aerosols above clouds from A-Train constellation coupled with SEVIRI. The detection algorithm encompasses a combination of spectral, textural, and temporal information from SEVIRI using Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) and Ozone Monitoring Instrument (OMI) as a benchmark. Aerosol properties from an AERONET station are used to create LUT for the spectral analysis. The uncertainty analysis is conducted based on user and producer accuracy.

3) Validate Spectrometers for Sky-Scanning, Sun-Tracking Atmospheric Research (4STAR) AOD measurements during NASA ORACLES aircraft field campaign with multispectral AOD retrievals from satellite sensors. The satellite retrieval involves a simultaneous retrieval of aerosol and cloud properties using color-ratio techniques in order to account for low COD biases due to overlying smoke aerosol absorption. Thereafter, document uncertainties of satellite retrieval based on variations of assumed aerosol absorption.
CHAPTER TWO

SEASONAL VARIATIONS OF THE ABOVE-CLOUD AEROSOL DIRECT RADIATIVE EFFECTS

2.1. Above-cloud aerosol radiative effects overview

The representation of aerosols, clouds, and aerosol-cloud radiative effects remains highly uncertain, restricting the reconstruction of past climate and the prediction of future climate change (Kahn, 2012; Myhre et al., 2013). The radiative effect due to scattering and absorption of shortwave (SW) radiation is known as the aerosol direct radiative effect (DRE) (Satheesh et al., 1999). The DRE in cloud-free regions generally leads to cooling at TOA and reduces surface temperatures since aerosols scatter more solar radiation than that of the ocean surface. The exception occurs over bright surfaces such as deserts (e.g. Patadia et al., 2009) and snow (e.g. Nair et al., 2013). Absorbing aerosols above clouds (AACs) also create a positive DRE at TOA since aerosols absorb cloud reflection (Chand et al., 2009; Feng & Christopher, 2015; Keil & Haywood, 2003; Meyer et al., 2013, 2015; Peers et al., 2015; Peters et al., 2011; Zhang et al., 2014, 2016). Additionally, the presence of AACs can lead to an underestimation of COT retrieval at visible wavelengths compared to the retrieval of COT in a pristine cloudy scene (Coddington et al., 2010; Haywood et al., 2004; Meyer et al., 2013). Moreover, absorbing aerosols near the surface can heat the atmospheric layer, raise the surface temperature, promote
atmospheric stability, and reduce relative humidity depending on the size, shape, and composition of aerosols (Kaufman et al., 2002).

Using CALIOP Aerosol and Cloud Layer Products and Moderate Resolution Imaging Spectroradiometer (MODIS) cloud product (MYD06), Meyer et al. (2013) found that the regional mean DRE efficiency in an AAC scene increases from 50.9 W m⁻² AOT⁻¹ to 65.1W m⁻² AOT⁻¹ after correcting for the low COT bias in the standard MODIS COT product at pixel-level computation. Although the pixel-level analysis provides a detailed DRE estimation, it requires high computational time and efforts. Subsequently, Zhang et al. (2014) suggested a more efficient method to calculate the DRE of AACs by using the joint histogram of cloud properties (i.e., COT and cloud top pressure) from the MODIS Level 3 cloud product and precomputed LUTs. They concluded that the change in COTs plays a greater role in the DRE than the change in above-cloud AOTs. While AACs generally lead to a positive DRE at TOA, the DRE varies significantly on daily, seasonal, and interannual scales according to measurements from Scanning Imaging Absorption Spectrometer For Atmospheric Chartography (SCIAMACHY) (de Graaf et al., 2014). Using eight years of CALIOP and MODIS observations, Zhang et al. (2016) found that the global ocean annual mean diurnally averaged AAC direct radiative effects are ~0.02 W m⁻², ~0.17 W m⁻², and ~0.15 W m⁻² at TOA, within the atmosphere, and at the surface, respectively. In the Southeast Atlantic, the annual mean diurnally averaged DREs are 0.21 W m⁻², 0.56 W m⁻², and -0.35 W m⁻² at TOA, within the atmosphere, and at the surface, respectively.

Multispectral analyses of absorbing aerosols above liquid clouds have been addressed in several studies. Jethva et al. (2013) introduced a color-ratio technique to simultaneously retrieve AOTs and COTs using the 470 nm and the 860 nm MODIS channels. They found that aerosols above clouds of COT < 2 (> 5) increase (reduce) reflectance at TOA. They also performed
sensitivity analyses on the AOT and the COT retrieval with respect to the single scattering albedo (SSA) and aerosol height biases. They found that an underestimation of the SSA leads to an underestimation of both AOTs and COTs while an underestimation of the aerosol height leads to an overestimation of optical thicknesses of both variables. The greatest overestimation occurs for AOT = 2, which is the thickest AOT examined in their study. Meyer et al. (2015) applied an optimal inversion method to simultaneously retrieve AOTs and underlying COTs and cloud effective radius (CER) using six MODIS channels ranging from visible to shortwave infrared wavelengths. Chang and Christopher (2016) adopted a color-ratio technique (Jethva et al., 2013) then added textural analysis and temporal information on SEVIRI to detect absorbing aerosols above closed-cell stratocumulus cloud. SEVIRI’s 15-minute temporal resolution is viable for tracking aerosols and underlying cloud and the daytime variation of their optical and radiative properties. A MODIS “Deep Blue” product dedicated to AAC retrieval is presently under development (Sayer et al., 2016), which will provide a significant contribution to AAC retrieval capabilities.

Using multi-angular information, total radiances, and polarized radiances from POLDER instrument, Peers et al. (2015) developed a technique to simultaneously retrieve aerosol and cloud properties for AAC scenarios, including AOT, COT, Angstrom exponent, and SSA. Over the Southeast Atlantic, the mean instantaneous AAC direct radiative effect during August 2006 was 33.5Wm$^{-2}$. The maximum instantaneous DRE of ~125Wm$^{-2}$ prevailed at 8°S near coastal regions. Using the Clouds and the Earth's Radiant Energy System (CERES) instrument and a RT model, Oh et al. (2013) documented that the global all-sky mean DRE increases with COT for COT > 2 but are weakened by both increasing surface albedo and CER.
Globally, the highest frequency of smoke above clouds occurs during months of June – August according to CALIOP observations (Devasthale & Thomas, 2011). The seasonal variability for the vertical separation of cloud and aerosol is greatest in the 0.0°–30.0°S latitude range. The largest vertical separation occurs within this latitudinal belt during September – November. Major contributions of aerosol in this region include biomass burning aerosols that are advected from Central Africa to the Southeast Atlantic and biomass burning aerosols that are advected from South America to the Eastern Pacific. Alfaro-Contreras et al. (2016) also studied AAC frequency on a global scale using measurements from both CALIOP and OMI. They also identified a high AAC frequency over the Southeast Atlantic from both sensors. The frequency of aerosol-cloud overlap increases from June until September and then decrease from September to October (Meyer et al., 2015; Zhang et al., 2016).

Min and Zhang (2014) examined the influence of cloud fraction on the diurnal cycle of the DRE using the rapid radiative transfer model, shortwave (RRTM_SW) model (Clough et al., 2005; Iacono et al., 2000). They noted that using an instantaneous cloud fraction data from MODIS-Aqua as a representation of daily mean cloud fraction led to an underestimation of cloud fractions and the DRE relative to a sinusoidal model. The sinusoidal model included a diurnal cycle of cloud fraction from SEVIRI and both MODIS instruments, providing a more realistic perspective of diurnal cloud fractions. Using the daily mean cloud fraction data from MODIS-Terra, in contrast, led to an overestimation of both cloud fractions and the DRE. Differences in cloud fractions (for low-level clouds) occurred because the peak cloud fraction typically took place at around 07:00 am local time and dropped to a minimum at around 05:00 pm. Thus, using a constant cloud fraction to assess its impact on DREs would introduce uncertainties associated the change in the solar zenith angle (SZA) and cloud fraction variability. Interestingly, they
found that the maximum DRE took place at around 09:00 am when both SZAs and cloud fractions are considered. They emphasized that using a grid mean COT to assess the DRE in lieu of a full histogram technique developed by Zhang et al. (2014) produced high biases since the former assumes plane-parallel cloud albedo below the aerosol layer. They also noted that an increase (decrease) in SSA led to a decrease (increase) in the DRE, confirming the importance of accounting for variations in aerosol properties when assessing the DRE of AACs.

During Southern African Fire-Atmosphere Research Initiative (SAFARI) in Year 2000, the C-130 aircraft conducted flights off coasts of Namibia and Angola. It was reported that the top and the base of biomass burning aerosols are located at about 5120 ± 550m and 1520 ± 660m, respectively (Haywood et al., 2004). During the flight on 7 September 2000, biomass burning aerosols were found to be located at altitudes between 1.8 – 3.7 km, whereas clouds are located below 1km (Keil & Haywood, 2003). Costantino and Bréon (2013) noted that it is not uncommon that absorbing aerosols and clouds coexist in a similar altitude in the Southeast Atlantic according to CALIOP observations, however. The frequency of this scenario increases westward away from the fire burning source. CALIOP can fail to identify aerosols below liquid clouds since clouds cause a complete attenuation of the backscattered radiation to the CALIOP sensor, resulting in undetected aerosols below liquid clouds (Winker et al., 2009; Young & Vaughan, 2009).

Johnson et al. (2004) simulated the direct and the semi-direct radiative effects of absorbing aerosols within and above clouds using the MET Office large eddy model. They found that AACs enhance the buoyancy of free-tropospheric air, thereby reducing the entrainment rate. Hence, the boundary layer becomes shallower and more humid, increasing the cloud liquid water path. Furthermore, the presence of above-cloud absorbing aerosols causes a reduction of the
downward flux reaching the cloud top, which is another contributing factor that enhances the cloud liquid water path. Both factors give rise to a negative semi-direct radiative effect. In contrast, the cloud liquid water path subsides when aerosols reside in the boundary layer because of the enhanced solar heating in the cloud layer. Aerosols aid to enhance the existing stability in the boundary layer during daytime, reducing the moisture flux from the surface to the cloud layer.

While previous studies have examined the diurnally averaged DRE of AACs from various observational approaches and modeling efforts to gain an insight on the radiative effects of AACs on climate, most studies have only briefly discussed the impact of SZAs on the DRE and the radiative heating rates (RHR) in AAC conditions. The objective of this study is to assess how variations of the SZA and vertical distributions of aerosols and clouds affect both DREs (at TOA, within the atmosphere, and at the surface) and RHRs in the lower troposphere on both monthly and diurnal scales. This work provides data and theoretical understanding to assist positioning science flights that aims at measurements of AAC radiative effects. This information is also useful for addressing the uncertainties of satellite observations under AAC conditions.

2.2. Data and Methodology

2.2.1. Satellite observations of AACs in the Southeast Atlantic

The Southeast Atlantic is dominated by the westward transport of high biomass burning aerosol loadings (Swap et al., 1996) and semi-permanent stratocumulus clouds during Austral winter and spring (June – October). This region, therefore, serves as a natural laboratory for studying above-cloud aerosols and their radiative effects (Alfaro-Contreras et al., 2016; Devasthale and Thomas, 2011; Meyer et al., 2013, 2015; Zhang et al., 2014, 2016). Nonetheless, there are challenges in simulating aerosol optical properties over Southeast Atlantic in global
aerosol models (Peers et al., 2016). An example of absorbing aerosols above clouds can be seen in Figure 2.1, which shows darkening of clouds caused by overlying aerosol absorption. The stronger attenuation of solar reflection at shorter wavelengths explains the changes in the cloud color observed in a true color composite from a multispectral sensor such as MODIS (e.g., Jethva et al., 2013, 2014, 2016).

![Figure 2.1](image.png)

Figure 2.1. MODIS-Aqua true-color composite (R=0.66 µm, G=0.55 µm, B=0.47 µm) on 2 August 2016. An example of aerosols above clouds is encapsulated in the blue polygon and that of pristine clouds are encapsulated within the red polygon.

### 2.2.2. Radiative transfer model description

To facilitate DRE assessments of AACs, the Discrete Ordinate Radiative Transfer (DISORT) (Stamnes et al., 1988) from the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al., 1998) is used. The uncertainty of flux calculations
is expected to be within 5% under clear-sky conditions (Gautier & Landsfeld, 1997). We mainly focus on the SW bands in this study since biomass burning smoke aerosol particles are primarily sensitive to the SW radiation (Haywood et al., 2003). The atmospheric conditions for the entire study are based on the default tropical atmospheric conditions of water vapor, temperature, and other atmospheric constituents (McClatchey et al., 1972). The ocean surface albedo from Tanre et al. (1990) is used to parameterize the surface spectral albedo. The surface albedos consist of 751 bands in the solar spectrum at 5 nm spectral resolution. The surface albedo is 0.03 at 0.5 µm.

2.2.3. Radiative transfer model experimental setup

Throughout this study, we use a set of representative aerosol and cloud parameters inferred from AAC climatology such as Meyer et al. (2013) and Meyer et al. (2015). Specifically, we apply a fixed AOT, COT, and CER of 0.6, 9.0, and 12.8 µm at 0.55 µm, respectively over selected locations in the Southeast Atlantic. In this study, we place aerosols and clouds between 2 – 4 km and 1 – 2 km above ground, respectively. These height values conform to the observations during SAFARI 2000 field experiments.

Aerosol properties including the spectral SSA, extinction efficiency, and asymmetry parameter that are used for the RT calculations follow those outlined in Meyer et al. (2015), which are based on aircraft measurements during the SAFARI 2000 field experiment (Haywood et al., 2003). We also compare radiative fluxes using SAFARI 2000 aerosol properties with those derived from the Level 2.0 AERONET data (Holben et al., 1998). One of the most frequently available AERONET data in Southern Africa is the Mongu, Zambia station. The aerosol properties from AERONET observation on 13 August 2006 are used as in Chang and Christopher (2016). The AERONET-derived SSA typically has an uncertainty of ~0.03 for AOT(0.44 µm) > 0.4 (Dubovik et al., 2000; Dubovik et al., 2002). In situ measurements during
SAFARI 2000 in both Mongu and Senanga of Zambia were lower by 0.02 than measurements taken by the respective AERONET stations (Leahy et al., 2007). Both the SSA and the asymmetry parameter of biomass burning aerosols decrease with wavelength, consistent with SAFARI 2000 data (Bergstrom et al., 2007). However, the SSA of aerosols gradually increases with time within a fresh biomass burning plume due to condensation of volatile organic compounds (Abel et al., 2003) and may increase with relative humidity as they are transported over the Southeast Atlantic (Haywood et al., 2003). The differences in the wavelength-dependent SSA of aerosols between the Southeast Atlantic and the Mongu station undoubtedly induce uncertainties when assessing DREs and RHRs.

2.3. Shortwave direct radiative effect calculations

The monthly diurnally averaged DRE of AACs is computed at three chosen locations (5.0°S, 15.0°S, and 25.0°S along 5.0°E) during which AACs are prevalent in Austral winter. It is computed by averaging the hourly instantaneous DRE, such that

$$\overline{DRE_{AAC,\text{month},\text{TOA}}} = \frac{1}{n} \sum_{i=1}^{n} \left[ (\downarrow F_{AAC,\text{TOA,inst},i} - \uparrow F_{AAC,\text{TOA,inst},i}) - (\downarrow F_{\text{cloudy,TOA,inst},i} - \uparrow F_{\text{cloudy,TOA,inst},i}) \right]$$

(2.1)

where $n$ is the number of observations during a month at a particular grid, $\downarrow F_{AAC,\text{TOA,inst},i}$ the instantaneous downward SW flux (W m$^{-2}$) at TOA at the $i$th hour for AACs, $\uparrow F_{AAC,\text{TOA,inst},i}$ the instantaneous upward SW flux for AACs at TOA, $\downarrow F_{\text{cloudy,TOA,inst},i}$ the instantaneous downward flux for pristine clouds at TOA, and $\uparrow F_{\text{cloudy,TOA,inst},i}$ the instantaneous upward flux for pristine
cloud at TOA. Since each day consists of twenty-four observations, the total number of observations in a month at a grid is the multiplication of twenty-four and the number of days in a month. Since $\downarrow F_{AAC,TOA,inst,i}$ is always equal to $\downarrow F_{cloudy,TOA,inst,i}$ at the same time, these two variables negate each other. Therefore, (2.1) can be simplified to:

$$\overline{DRE_{AAC,\text{month},TOA}} = \frac{1}{n} \sum_{i=1}^{n} (\uparrow F_{cloudy,TOA,inst,i} - \uparrow F_{AAC,TOA,inst,i})$$

(2.2)

The monthly diurnally averaged DRE at the surface (SFC) for each month is given by

$$\overline{DRE_{AAC,\text{month},SFC}} = \frac{1}{n} \sum_{i=1}^{n} \left[ (\downarrow F_{AAC,SFC,inst,i} - \uparrow F_{AAC,SFC,inst,i}) - (\downarrow F_{cloudy,SFC,inst,i} - \uparrow F_{cloudy,SFC,inst,i}) \right]$$

(2.3)

Hereinafter, Equation (2.3) cannot be simplified since $\downarrow F_{AAC,SFC,inst,i}$ is not equal to $\downarrow F_{cloudy,SFC,inst,i}$. The solar radiation that reaches the surface is lower in an AAC case than in a pristine cloudy case. The monthly mean DRE within the atmosphere is the difference between (2.2) and (2.3).

2.4. Shortwave radiative heating rate calculations

Vertical profiles of the instantaneous RHR provide information on the magnitude of radiative warming/cooling of an atmospheric layer (i.e., between two atmospheric levels). The flux portion of the RHR equation is obtained by subtracting the sum of the downward flux at the
lower pressure level and the upward flux at the upper pressure level from the sum of the
downward flux at the upper pressure level and the upward flux at the lower pressure level. The
equation for the instantaneous shortwave RHR (e.g., Guan et al., 2010; Quijano et al., 2000) at a
layer is given by

$$\frac{\partial T}{\partial t_{AAC,inst}} = \frac{g}{c} \left( \frac{\downarrow F_j - \downarrow F_{j+1} + \uparrow F_{j+1} - \uparrow F_j}{|p_j - p_{j+1}|} \right)$$

(2.4)

where $g$ is acceleration due to gravity, $c$ the specific heat capacity of dry air at constant pressure,
$P_j$ the upper (denoted by “$j$”) pressure level (Pascal), $P_{j+1}$ the lower (denoted by “$j+1$”) pressure
level, $\downarrow F_j$ the downward flux at the upper pressure level, $\downarrow F_{j+1}$ the downward flux at the lower
pressure level, $\uparrow F_{j+1}$ the upward flux at the lower pressure level, and $\uparrow F_j$ the upward flux at the
upper pressure level. The raw unit for RHR is expressed as Kelvin per second but are generally
converted to Kelvin per day. The mean RHRs over a given time period in an atmospheric layer
are expressed as follows:

$$\frac{\partial T}{\partial t_{AAC,n}} = \frac{1}{n} \frac{g}{c pd} \sum_{l=1}^{n} \left[ \frac{(\downarrow F_{l,j} - \downarrow F_{l,j+1} + \uparrow F_{l,j+1} - \uparrow F_{l,j})}{|p_{l,j} - p_{l,j+1}|} \right]$$

(2.5)

Vertical distributions of aerosols can significantly impact RHR since they warm the
atmosphere at which they reside by absorbing solar radiation (Guan et al., 2010; Liao & Seinfeld,
1998). In the Southeast Atlantic, aerosol vertical distributions can vary in both cloudy and cloud-
free conditions as seen in two cases from CALIOP in Figure 2.2. The base of aerosols could be
either vertically spread throughout the atmospheric column (Figure 2.2a) or have an observable gap from underlying stratocumulus clouds (Figure 2.2c). Between 6.3°S to 6.5°S in the first scenario (Figure 2.2a), aerosols vertically extend from the surface to ~4 km. The aerosol subtype layer information in Figure 2.2b indicates that smoke aerosols dominate this domain. Between 5.8°S to 6.2°S, clouds occur at ~1.2 km above the surface with an attenuation underneath this altitude. Whether aerosols are present below the cloud layer are unknown due to cloud attenuation. Since a narrow smoke layer is present from the surface up to ~0.8 km at 5.7°S, one could speculate that aerosols may be present below the cloud layer that is bounded by the adjacent smoke layers. The second scenario highlights an AAC case on 13 August 2006 (Figure 2.2c). Aerosols from 19.0°S to 20.0°S clearly reside above clouds, but various aerosol types are present between these two latitudes as shown in the aerosol subtype layer (Figure 2.2d). Liu et al. (2015), however, argued that the frequency of aerosol mixing may be overestimated by the aerosol subtype classification algorithm in the Southeast Atlantic during which the biomass burning season prevails.
Figure 2.2. (a) The CALIOP VFM feature layers for 19 August 2009 at 1339Z. (b) The aerosol subtype layers for the identical time and location as (a). (c) As in (a), but for 13 August 2006 at 1323Z. (d) The aerosol subtype layers for the identical time and location as (c).
2.5. Shortwave AAC radiative effects and radiative heating rates

2.5.1. Impact of vertical distributions

Diurnally averaged upward TOA fluxes, downward fluxes between 0 – 4 km, and RHRs between 0 – 4 km for SZAs based on 1 September 2016 at 15.0°S 5.0°E for eight different vertical distributions of aerosols and clouds are shown in Figure 2.3. These cases are chosen to represent the major scenarios in estimating the aerosol DREs. RHRs are calculated on an hourly basis using Equation (2.5). Each scenario is represented by a circled alphabet. Note that the date and location used in Figure 2.3 do not necessarily reflect the actual aerosol properties and cloud properties on this day at this location. Aerosol optical properties are either based on the SAFARI 2000 field experiment or the AERONET observation in Mongu. For a scenario that contains aerosols (i.e., Scenarios B to G), the total column AOT is 0.6 and is distributed evenly within the designated range of altitudes by 1 km increment. For instance, the AOT in Scenarios B, C, F, and G, is 0.3 in each atmospheric layer. The AOT, in contrast, is only 0.15 in each atmospheric layer in Scenarios D and E. Discussions of radiative fluxes and RHRs for each scenario are as follows:

A. Clear-sky - The reduction of downward fluxes from TOA towards the Earth’s surface results primarily from molecular scattering and gaseous absorption. A typical tropical atmospheric condition enhances these effects more than other types of atmospheric profile assumptions. Thus, other atmospheric profiles would have a higher surface flux but lower RHRs due to weaker gaseous absorption. The upward TOA flux of 28 W m⁻² comprises atmospheric and surface scattering. RHRs in the lowest 4 km range between 0.8 and 0.9 K day⁻¹.
Figure 2.3. A conceptual diagram showing diurnally averaged radiative fluxes (in W m$^{-2}$) and RHRs (in K day$^{-1}$) for eight scenarios (circled alphabets) from the RT model for SZAs based on 1 September 2016 at 15.0°S 5.0°E. The TOA downward flux in each scenario is ~396 W m$^{-2}$. Yellow boxes represent locations of aerosol layers based on the SAFARI 2000 aerosol model. Brown boxes denote locations of aerosol layers based on the Mongu AERONET observation on 13 August 2006 (only elevated aerosol cases are shown). Light blue boxes denote cloud layers. An overlap of cloud and aerosol takes place in Scenario E between 1 km and 2 km. Values encapsulated in blue arrowed boxes denote the upward TOA fluxes. The purple horizontal bar at the bottom of the diagram represents the ocean surface. Values in black between 0 km – 4 km within a layer represent RHRs. Each value in red adjacent to the downward red arrow represents the downward flux entering the underlying atmospheric layer. The AOT, COT, and CER for all cases are 0.6, 9.0, and 12.8 µm at 0.55 µm, respectively. All flux values are rounded off to the nearest whole W m$^{-2}$. RHRs are rounded off to a tenth of K day$^{-1}$. Note that the schematic is not presented in scale.
B. Cloud-free elevated aerosols (Mongu AERONET)- The presence of AOT = 0.6 between 2 – 4 km (i.e., AOT of 0.3 within each atmospheric layer) using the Mongu AERONET aerosol optical properties leads to relatively lower downward surface fluxes compared to the clear-sky scenario due to an additional absorption and reflection by the aerosol layer. The aerosol layer enhances the upward TOA flux as a result of an enhanced aerosol scattering. The RHR increases by over a factor of three within the aerosol layer compared to the clear-sky scene.

C. Cloud-free elevated aerosols (SAFARI 2000)- When substituting the aerosol optical properties in Scenario B with the SAFARI 2000 aerosol optical properties (less absorbing than Mongu AERONET aerosol optical properties) while fixing the total column AOT, both TOA and surface fluxes increase. This phenomenon occurs since the aerosol optical properties in this scenario has a weaker absorption efficiency (i.e., a higher SSA). The RHR is almost a factor of two smaller than that in Scenario B in the aerosol layer.

D. Cloud-free aerosols- The aerosol extinction is evenly distributed between 0 – 4 km in this case, so the AOT within each atmospheric layer is 0.15. Thus, the downward flux in each atmospheric layer is relatively higher than those in Scenario C. The RHR in each layer is also lower between 2 – 4 km because each layer consists of a lower AOT. However, both the downward surface flux and the upward TOA flux remain identical to Scenario C, indicating that the vertical distribution of aerosols mainly affects RHRs of atmospheric layers.

E. Cloud-embedded aerosols- A cloud layer embedded in aerosols between 1 – 2 km with COT of 9.0 causes an enhancement to the upward TOA flux and a reduction in the downward surface
flux compared to Scenario D. Note that aerosol-cloud microphysical interactions (i.e., aerosol indirect effects) are not accounted for in the RT model, so the computation is solely based on scattering and absorption of aerosols and clouds. Clouds also elevate the RHR in the overlying aerosol layers due to aerosol absorption of scattered radiation from cloud-top. In contrast, the aerosol layer below the cloud layer experiences a reduction in RHR compared to the scenario without clouds. The diurnally averaged downward flux at cloud top increases from 292 W m\(^{-2}\) to 304 W m\(^{-2}\) from Scenario D to Scenario E. This increase results from scattering of the aerosol base above cloud-top.

F. Elevated aerosols above clouds (SAFARI 2000)- The upward TOA flux is lower than that in Scenario E since the total AOT above cloud top is higher in these cases. Nevertheless, the downward surface fluxes are identical in both scenarios, suggesting that downward surface fluxes are primarily determined by the total column AOT and COT rather than their differential vertical distributions. Even when elevated aerosols are located at the upper troposphere (e.g., 8 km), both TOA and surface fluxes only increase by less than 1 W m\(^{-2}\) (not shown). The slight increase in the TOA flux is due to the reflection of higher flux at higher altitude where less molecular scattering and absorption had taken place. Thus, molecular scattering and absorption play relatively minor roles in TOA fluxes when aerosols and clouds are optically thick.

G. Elevated aerosols above clouds (Mongu AERONET)- Both upward TOA and downward surface fluxes decrease when the elevated aerosol layer above cloud consists of strong absorbing aerosols instead of light absorbing aerosols. The RHR in the aerosol layer increases when switching the aerosol optical properties from SAFARI 2000 to the Mongu AERONET. Likewise,
the diurnally averaged TOA flux decreases from 137 to 120 W m$^{-2}$ after altering the aerosol model. The aerosol layer experiences a higher RHR than cloud-free elevated aerosol case (Scenario B) since the reflection of the cloud-top exceeds the reflection of the ocean. Therefore, the RHR at this layer is relatively higher than the case without underlying clouds despite having the same AOT as that in Scenario B. The higher absorption of Mongu AERONET aerosol optical properties than those from SAFARI 2000 results in a decrease of downward flux at cloud-top from 285 W m$^{-2}$ to 262 W m$^{-2}$.

H. Cloud only- This scenario has the highest upward TOA flux among the eight scenarios due to the absence of solar absorption by overlying aerosols. The downward flux at cloud-top is 324 W m$^{-2}$. The RHR in the cloud layer is 1.8 K day$^{-1}$, which is 0.1 K day$^{-1}$ and 0.2 K day$^{-1}$ higher than Scenarios F and G, respectively. These differences suggest that overlying optically-thick and absorbing aerosols reduce the solar heating in the cloud layer.

2.5.2. Dependence of DREs on SZA

DREs as a function of SZA based on aerosol and cloud optical properties in Scenario F of Figure 2.3 are illustrated in Figure 2.4. The DRE at TOA (Figure 2.4a) and within the atmosphere (Figure 2.4b) are positive while the DRE at the surface (Figure 2.4c) is negative, which are consistent with the signs of DREs estimated by Zhang et al. (2016) in the Southeast Atlantic. The TOA experiences a positive DRE because upward TOA fluxes are higher for thick pristine clouds than for absorbing aerosols above. The DRE at TOA increases with SZA from 0° to 54°. The peak DRE at TOA takes place at SZA = 54°, with a value of ~29 W m$^{-2}$, where the pristine cloudy flux and the AAC flux at TOA are ~330 W m$^{-2}$ and ~301 W m$^{-2}$, respectively. A decrease of the DRE with an increased SZA occurs when SZA > 54°, potentially yielding the
same DRE for different SZAs when SZA < 76°. The surface DRE increases from ~71 W m⁻² to ~0 W m⁻² when the SZA increases from 0° to 90°. Within the atmosphere, the DRE decreases from ~89 W m⁻² to 0 W m⁻² with an increase of SZA from 0° to 90°. The DRE within the atmosphere responds greater to changes in SZA than that at TOA since the surface DRE responds greatly to the changes in SZA.

![Graphs showing DREs as a function of SZA](image)

Figure 2.4. DREs as a function of SZA (a) at top-of-atmosphere, (b) within the atmosphere, and (c) at the surface based on aerosol and cloud properties in Scenario F of Figure 2.3.

2.5.3. Seasonal variations of DRE

Monthly diurnally averaged DREs at TOA, within the atmosphere, and at the surface at 5.0°S, 15.0°S, and 25.0°S along 5.0°E are presented in Figure 2.5. These computations follow Scenario F of Figure 3, which are elevated aerosols above clouds based on SAFARI 2000 aerosol optical properties. The DRE at TOA among the three locations varies between 10 – 11 W m⁻², with a decreasing trend with month at 5.0°S and an increasing trend with month at 15.0°S and 25.0°S. This minor seasonal change in the DRE at TOA results from a range of only ~18.3 W m⁻² to ~29 W m⁻² for SZA < 76°. At noon, the DRE at TOA is ~18.9 W m⁻², ~20.5 W m⁻², and ~23.1 W m⁻² at 5.0°S, 15.0°S, and 25.0°S along 5.0°E, respectively. The increasing DRE with latitude is expected since the DRE increases with SZA when SZA < 54°. Off coastal Angola, the local seasonal mean DRE occasionally exceeds 30 W m⁻² according to satellite measurements.
(Peters et al., 2011). The surface DRE responds greater to changes in SZA as anticipated based on Figure 2.5c. At 5.0°S 5.0°E, it decreases from ~-18.7 W m⁻² to ~-22.0 W m⁻² from June to October. During the same period, the surface DRE decreases from ~-12.2 W m⁻² to ~-22.1 W m⁻² at 25.0°S 5.0°E since the seasonal variation of SZA is greater at this location than that at 5.0°S 5.0°E. The surface DRE among the three locations nearly converge at -22 W m⁻² in October since the SZA among the three locations are almost equal as the month progresses from June to October. Within the atmosphere, this convergence can also be observed at ~33 W m⁻² in October.

Figure 2.5. The monthly mean (June – October) diurnally averaged DRE (in W m⁻²) at top-of-atmosphere, within the atmosphere, and at the surface at 5.0°S (red), 15.0°S (green), and 25.0°S (blue) along 5.0°E, respectively. Aerosol and cloud properties follow those described in Scenario F.

2.5.4. Diurnal variations of DRE

Diurnal variations of DREs at TOA, within the atmosphere, and at the surface at the three selected locations (5.0°S, 15.0°S, and 25.0°S along 5.0°E) are shown for 1 September 2016 excluding hours during which SZA exceed 90° (Figure 2.6). At 5.0°S 5.0°E, the primary peak DRE at TOA (~29.5 W m⁻²) occurs at 8Z while the secondary peak DRE at TOA (~29.4 W m⁻²)
takes place at 15Z. At 25.0°S 5.0°E, the primary peak DRE (~29.4 W m$^{-2}$) at TOA occurs at 15Z while the secondary peak (~28.9 W m$^{-2}$) occurs at 9Z. These trends are consistent with the fact that the DRE increases from early morning hours, reaching a peak between sunrise and noon when SZA = 54°. The surface DRE reaches its minimum at 12Z as expected. An equatorward decrease of the surface DRE can be observed as a result of a decreasing SZA. Field experiments that are dedicated to a complete diurnal cycle of AACs would provide an insightful comparison with the results presented here.

2.6. Hourly variations of radiative heating rates and radiative fluxes

Monthly mean hourly variations of RHRs from June to October in the lower troposphere (0 – 4 km) based on aerosol and cloud properties in Scenario E of Figure 2.3 are shown in Figure 2.7. This figure also shows monthly mean downward TOA fluxes, monthly mean downward fluxes at cloud-top, and monthly mean RHR differences (pristine clouds minus AACs) in the cloud layer. As in Figure 2.3, the geographic coordinate at 15.0°S 5.0°E serves as a representative location. The peak downward TOA fluxes occur at 13Z, which is an hour after the peak DRE at TOA. As the SZA decreases with month, the difference in the downward flux at cloud-top between pristine clouds and AACs becomes smaller. In June at 13Z for example, the downward flux at cloud-top decreases from 889 W m$^{-2}$ in a pristine cloudy condition to 849 W m$^{-2}$ in an AAC condition. In October at 13Z, the downward flux at cloud-top decreases from 1146 W m$^{-2}$ to 1113 W m$^{-2}$ for the same change. Hence, the difference between these two conditions during peak hours drops from 50 W m$^{-2}$ to 33 W m$^{-2}$ between June and October.
Figure 2.6. The diurnal variation of DRE at top-of-atmosphere, within the atmosphere, and at the surface at 5.0°S (red), 15.0°S (green), and 25.0°S (blue) along 5.0°E during 1 September 2016. Only the hours where SZA < 90° are shown. Aerosol and cloud properties follow those described in Scenario F.
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Figure 2.7. Monthly mean diurnal variations of RHRs (in Kelvin per day) between 0 – 4 km based on aerosol and cloud properties in Scenario E of Figure 2.3 and downward TOA fluxes in W m\(^{-2}\) (red text adjacent to the red down arrow) at 15.0°S 5.0°E between June – October 2016. The change in RHRs in the cloud layer is shown between 1 – 2 km where positive values denote Scenario E minus Scenario H and values in parentheses denote Scenario F minus Scenario H. Vertical numbers between 3 – 4 km denote the downward fluxes at cloud-top for Scenarios H, E, and F (left to right). Only the hours where SZA < 90° are shown. Aerosols and clouds are located between 0 – 4 km and 1 – 2 km above the ground, respectively. Note that a truncation in altitude occurs between TOA and 4 km.

Contrary to peak hours, the difference in the downward flux at cloud-top in the early morning hour (i.e., 8Z) between the pristine cloudy and the AAC condition increases with month. In June at 8Z, the downward flux at cloud-top decreases from 198 W m\(^{-2}\) to 156 W m\(^{-2}\) (42 W m\(^{-2}\) difference). In October at 8Z, the downward flux at cloud-top decreases from 388 W m\(^{-2}\) to 338 W m\(^{-2}\) (50 W m\(^{-2}\) difference). Likewise, this monthly increasing trend also takes place late in the afternoon (i.e., 16Z). These findings may suggest that aerosols play a greater radiative role in the diurnal cycle at cloud-top later in the season during early morning/late afternoon hours and earlier in the season during midday.

The RHR for elevated above-cloud aerosols (Scenario F) with AOT = 0.6 shows a difference of -0.2 K day\(^{-1}\) compared to the pristine cloud case (Scenario H) between 10 – 16 Z in June. By October, the -0.2 K day\(^{-1}\) difference spans between 9 – 17 Z. A lower downward flux at
cloud-top for these cases than that for cloud-embedded aerosols (Scenario E) is not surprising since the AOT above cloud in Scenario E is only 0.3. The reduction of the total (shortwave and longwave) RHR supports previous studies that the rate of cloud fraction decreases at a slower rate during daytime for aerosols above clouds, which may partly explain the weak diurnal cycle of cloud fraction in the Southeast Atlantic (Burleyson & Yuter, 2015a). Moreover, the diurnal variations of liquid water path, cloud-top height, and cloud geometrical thickness in the Southeast Atlantic tends to be weaker than those in the Southeast Pacific (Painemal et al., 2015). Whether the finding of these studies depend on the characteristics of overlying aerosols require further investigation.

The increase of RHR in the cloud layer with month is also seen in Figure 2.7. In June and July, the RHR reaches ~6.6 K day$^{-1}$ at noon within the cloud layer (1 – 2 km). By October, the RHR reaches ~8.9 K day$^{-1}$. The differences in RHR between the cloud-embedded aerosol condition (Scenario E) and the pristine cloudy condition also increases with season, especially near midday. For example, a difference of 1.3 K day$^{-1}$ is observed in June at 12Z and 13Z, increasing to 1.7 K day$^{-1}$ in October. Thus, aerosols that coexist in a cloud layer enhance the RHR, which would promote a thinning of the cloud geometric thickness and a reduction of the COT. This process links to the positive semi-direct radiative effects (e.g., Johnson et al., 2004).

Satellite and ship observations have shown that stratocumulus cloud fraction tends to peak before dawn and reaches a minimum around sunset on the eastern portion of major oceans in the sub-tropical anticyclonic belt (Burleyson & Yuter, 2015a, 2015b). As a result, the weakening of solar heating should reduce the dissipation of underlying clouds than no aerosols above clouds (Johnson et al., 2004; Wilcox, 2010, 2012). Since the sustenance of stratocumulus clouds depends on the magnitude of the total RHRs at cloud top, the SW heating by overlying
aerosols acts to reduce the total RHR at cloud-top. Moreover, the overlying aerosols strengthen the stability of the inversion layer (i.e., entrainment interfacial layer), which increase the lower-tropospheric stability. These processes allow moisture to be confined in the boundary layer (Johnson et al., 2004; Wood, 2012).
CHAPTER THREE

IDENTIFYING ABSORBING AEROSOLS ABOVE CLOUDS FROM SEVIRI

3.1. Retrieval overview

The method of aerosol absorption detection using satellite UV observations was developed by P.K. Bhartia at the NASA Goddard Space Flight Center (Torres & Remer, 2013) and first documented by Hsu et al. (1996) using reflectivity differences between two near-UV channels from the Nimbus-7 Total Ozone Mapping Spectrometer (TOMS). Quantification of aerosol absorption has been conducted by Torres et al. (1998) in a theoretical framework using the “residual method” based on the departure of the observed spectral differences in near-UV radiances from that of a molecular atmosphere. Radiative effects of smoke aerosols above clouds over southeast Asia using UV Aerosol Index (AI) as an indicator of smoke presence above cloud decks have been performed (Hsu et al., 2003).

Inversion approaches from polarization measurements provide the capability for retrieving above-cloud AOTs. A novel strategy for retrieving above-cloud fine mode AOTs using polarized phase function at forward scattering angles from the POLDER has been pioneered (Waquet et al., 2013). Simultaneous retrieval of AOT and COT has been conducted using inversion methods from near-UV radiances in OMI (Torres et al., 2012) and from
visible/near-infrared color ratio techniques in MODIS (Jethva et al., 2013). Such an approach has also been applied to simultaneously retrieve the aerosol absorption optical thickness and the underlying COT from POLDER measurements (Peers et al., 2015). Meyer et al. (2013) performed corrections on low COT bias from overlying absorbing aerosols for August and September between 2006 – 2011 over the southeast Atlantic. They noted that adjusting for above-cloud aerosol attenuation at 0.86µm increases the regional mean COT by ~6 % relative to the existing standard MODIS cloud product (i.e., MOD06 and MYD06). Meyer et al. (2015) developed a technique to simultaneously retrieve above-cloud AOT and underlying liquid cloud optical and microphysical properties by utilizing six MODIS channels ranging from VIS to shortwave infrared wavelengths. Their LUT covered AOT, COT, and \( \tau \), using an optimal estimation method. Various AOT retrieval approaches have been introduced on CALIOP such as the extinction to backscatter ratio technique (Young & Vaughan, 2009), the color ratio technique (Chand et al., 2008), and the depolarization ratio approach (Yu et al., 2015). Inter-sensor comparison among these sensors has revealed consistent above-cloud AOT retrievals. However, the CALIOP 532nm have shown to underestimate the above-cloud AOT due the solar background illumination, causing a lower signal-to-noise ratio during daytime retrieval (Liu et al., 2015; Peers et al., 2015).

Multi-sensor applications have also been performed in several studies. Ricardo Alfaro-Contreras et al. (2014) assessed the low COT bias due to smoke aerosols above clouds by collocating the OMI AI, CALIOP, and MODIS. They further compared the standard MODIS COT, derived from the 0.86 μm channel, with the MODIS supplementary product (derived mainly from the shortwave-infrared 1.64 μm channel). Both products were then referenced to COT at 0.646μm. Since the VIS channel is more sensitive to smoke attenuation than the near-
infrared (NIR) channel, the retrieved COT differences between these two channels yield the bias associated with smoke above clouds. They indicated that for OMI AI exceeding 1, the low COT bias ranges between 10 – 20 % at 0.86 µm for smoke above clouds over the south Atlantic. A negative semi-direct radiative forcing results in a thickening of stratocumulus clouds due to overlying smoke aerosols based on A-Train measurements (Wilcox, 2010). Wilcox (2012) found a net positive radiative forcing of 0.3 Wm⁻² for smoke above clouds between the direct and the semi-direct radiative forcing. The direct radiative effects of absorbing aerosols above clouds are also sensitive to the optical thickness of absorbing aerosols. Zhang et al. (2014) implemented a technique to estimate absorbing (smoke and polluted dust) aerosols above clouds direct radiative effect using CALIOP and MODIS. They found an direct radiative effect of ~30.9 Wm⁻² after making an adjustment to the CALIOP AOT’s low bias by a factor of 5 (Jethva et al., 2014).

The aforementioned studies have improved our understanding of above-cloud aerosol radiative effects. However, the temporal resolution of A-Train satellites are limited to one daytime overpass over a particular location per day, thereby inhibiting aerosols above clouds studies on a sub-daily scale. While CALIOP has enabled a vertical viewing capability of aerosols above clouds, one of its major shortcomings pertains to its near-nadir viewing restrictions with a 16-day repeat cycle (Winker et al., 2009). Such limitations hamper an extensive horizontal spatial analyses of aerosols above clouds. Given the high temporal evolution in the properties of aerosols (Takemura et al., 2002), monitoring the diurnal variation of the presence of aerosols above clouds will advance our understanding of short-term aerosols above clouds characteristics. Over the southeast Atlantic, SEVIRI aboard Meteosat Second Generation (MSG) satellite, located at the prime meridian above the equator, serves as the geostationary satellite platform for this region (Schmetz et al., 2002).
Numerous studies have utilized SEVIRI for feature detection that relies heavily on the high temporal resolution satellite data. For example, Derrien & Gléau (2010) implemented a temporal differencing technique to identify clouds at high solar zenith angles (during sunset and sunrise). SEVIRI’s high temporal resolution has also provided capabilities for monitoring dust storm outbreaks in the Mediterranean region and the Arabian Peninsula (Sannazzaro et al., 2014), detecting and tracking volcanic ash (Christopher et al., 2012; Naeger & Christopher, 2014), and developing a fire detection algorithm over Africa (Roberts & Wooster, 2008). The present study develops a method to identify smoke above closed-cell stratocumulus clouds (Sc) using SEVIRI’s multispectral, textural, and temporal characteristics. The condition that mostly resembles plane-parallel approximation in radiative transfer calculations is the extensive coverage of closed-cell Sc, so accurately identifying this feature is critical. The algorithm can be used to study the diurnal variation of smoke above Sc at a high temporal resolution. To keep the scope manageable, this study only concentrates on the algorithm development of smoke above closed-cell Sc detection.

3.2. Study area

An example of absorbing aerosols above clouds over the southeast Atlantic from different sensors is shown in Figure 3.1. Figure 3.1a shows a RGB three-band overlay of a SEVIRI image on 13 August 2006 at 1330Z over the southeast Atlantic Ocean. The central wavelengths of the red channel, the green channel, and the blue channel are 1.64 μm, 0.81 μm, and 0.64 μm, respectively. Yellow arrows depict wind vectors at 850 hPa, which is the nearest pressure level from the cloud top pressure. The maximum wind speed (10.2 ms⁻¹) corresponds to the longest vector on this image, which is located at the bottom right of this image. Pristine clouds appear as white since they are spectrally flat given their non-selective absorbing nature (Wang & Shi,
The light bluish regions are either cloud edges or thin clouds due to lower NIR reflectance at cloud edges (Nakajima & King, 1990). The tannish discoloration of clouds occurs as a result of smoke attenuation in the green channel and the blue channel (Alfaro-Contreras et al., 2014; Jethva et al., 2013).

The southeast Atlantic is dominated by high biomass burning aerosol loadings and semi-permanent Sc during Austral winter (June – September) (Wilcox, 2010, 2012; Yu et al., 2012). The algorithm in this study is specifically designed for detecting smoke above closed-cell Sc, which is characterized with ascending motion at the cell center and descending motion at the edge of the cell. This cloud type mostly appears over the eastern portion of ocean basins where a relatively cooler ocean current prevails. On the contrary, open-cell Sc is dominated by descending motion at cell-center and ascending motion at their edges (Agee et al., 1973; Stevens et al., 2005). The large contrast in both reflectance and temperature properties in open-cellular convective regions results in high uncertainty for their detection as opposed to a more homogenous reflectance and temperature associated with closed-cell Sc. While this study focuses on aerosols originating from biomass burning in southern Africa, aerosols from South America could be transported eastward (Bachmeier & Fuelberg, 1996; Talbot et al., 1996) to cause potential intercontinental aerosol mixing. However, the frequency of such a mixing above liquid clouds in the southeast Atlantic remains uncertain (Meyer et al., 2015). The remainder of this study will focus on detecting smoke aerosols (originated from southern Africa) above clouds associated with closed-cellular convection.
Figure 3.1. (a) SEVIRI with a CALIOP overpass in green and red on 13 August 2006. Yellow arrows represent wind vectors at 850 hPa. (b) OMI UV Aerosol Index at 1315 Z on this day. The black (white) line represents the CALIOP overpass along the green (red) line in (a). The pink lines represent CALIOP overpasses that were used for uncertainty analysis for the selected case days. (c) VFM feature layers from CALIOP. (d) Aerosol subtype layers from CALIOP. In both (c) and (d), green and red transects correspond to the CALIOP overpass in (a).
3.3. Data and methodology

The A-Train data sets are coupled with SEVIRI (3-km nadir resolution) (Schmetz et al., 2002) to leverage an algorithm for detecting smoke above closed-cell Sc. This imager consists of eleven spectral channels (three solar channels and eight thermal channels) and a broadband high-resolution visible channel. The algorithm undergoes three major processes at a pixel-level. We begin by using spectral thresholds to screen for smoke above low-level liquid clouds. Next, we perform a textural analysis to screen for cloud center and cloud edges. Finally, we apply temporal tests to screen for close-cell Sc. The eventual algorithm aims to identify smoke above closed-cell Sc at a pixel-level. A-Train data including OMI AI and CALIOP lidar information will serve as a benchmark for the algorithm development. The UV AI data are obtained from Version 3 of OMI/Aura Level 2 near-UV Aerosol data product (OMAERUV_V003). This index is calculated from the residual quantity based on the logarithm of the ratio of the measured radiances to the model-calculated radiances for a molecular-exclusive atmosphere. It has a spatial resolution of 13 km × 24 km at nadir (Torres et al., 2012). Given the high uncertainty of identifying aerosols with low AI values (Torres et al., 1998), only AI values above 0.5 are identified as absorbing aerosols in this study. This threshold has also been adopted in Yu et al. (2012) for above-cloud AOT assessment and in Feng and Christopher (Feng & Christopher, 2015) for studying radiative effects of absorbing aerosols above clouds. Figure 3.1b illustrates OMI AI for 13 August 2006 over the southeast Atlantic. Strong UV absorption can be seen at the center of this image as indicated by AI values exceeding 4. A high sensitivity between the AI and the aerosol-cloud separation distance, particularly for absorbing aerosols above thin clouds with high separation distance (Torres et al., 2012). Another uncertainty in the interpretation of AI arises from the wavelength-dependent absorption AOT attributed to the aerosol single scattering.
albedo and the Angström absorption exponent. Note that this study merely focuses on detecting the presence of smoke aerosols above clouds. We use Level 2 Version 3.01 cloud layer and aerosol layer detection products at 5 km horizontal resolution (Winker et al., 2009). Additionally, the CALIOP Level 2 Vertical Feature Mask (VFM) and the aerosol subtype (Omar et al., 2009) have been used to screen for columns that exclusively contain smoke above clouds within each of the 5 km cloud and aerosol layer horizontal domain. The VFM and the aerosol subtype have a horizontal resolution of ~333 m and a vertical resolution of 30 m between altitudes of 0.5 km and 8.2 km. Only a high confidence of aerosol and cloud detection is considered in this study. To eliminate possible mixings between aerosols and clouds within a vertical column, we enforce a 300 m minimum vertical separation distance between the smoke base and the cloud top. Since the standard VFM is generated from the 532 nm channel, the geometrical thickness of smoke layer tends to be underestimated during daytime due to solar background illumination (Liu et al., 2015). The underestimation of geometrical thickness of smoke suggests that the smoke base may be much closer to the cloud top than the smoke base indicated by the VFM data. Using the VFM derived from the 1064 nm channel may be an alternative solution to detecting thin aerosols. Ultimately, the goal is to confirm the presence of smoke above clouds from SEVIRI when CALIOP identifies the same feature. Figure 3.1c shows the CALIOP feature layer and Figure 3.1d shows the aerosol subtype along the green line and the red line in Figure 3.1a. Since this study focuses on identifying smoke above clouds, we only select the aerosols that consist of only smoke aerosols within the entire 5 km aerosol feature layer.

The Aqua-MODIS Level 2 Collection 6 cloud product (MYD06) at 1 km nadir resolution is used to obtain COT and CER (Platnick et al., 2003) along the CALIOP overpass. While the OMI footprint exceeds any given MODIS footprint, we collocated the OMI pixel closest to the
MODIS cloud pixel to obtain the aerosol absorption strength. Thus, an OMI footprint potentially serves as a collocation pixel for multiple SEVIRI, CALIOP, and MODIS pixels. Note that the present study aims to develop an algorithm for detecting smoke above closed-cell Sc on SEVIRI rather than deriving an algorithm for detecting aerosols of various AOTs above clouds of various COTs. However, an LUT for simultaneous AOT and COT retrievals for smoke above clouds is created for developing spectral thresholds. Low COT biases due to overlying smoke attenuation in the MYD06 product means that the minimum COT of this algorithm is likely the absolute minimum COT value that is valid for user applications. The presence of smoke above clouds does not cause a significant CER retrieval bias since its retrieval depends on shortwave infrared wavelength (i.e., 1.64 µm), which is mostly transparent to submicron smoke aerosols (Ricardo Alfaro-Contreras et al., 2014; Meyer et al., 2015).

3.3.1. Radiative transfer calculations

To guide the selection of spectral thresholds in the SEVIRI algorithm, simultaneous retrievals of AOT and COT is performed using SBDART (Ricchiazzi et al., 1998). Details of the input parameters in the LUT are outlined in Table 3.1. Input aerosol properties follow the AERONET Level 2.0 observations at Mongu, Zambia on 13 August 2006, which are based on bimodal lognormal size distributions (Dubovik et al., 2000). Combining the particle size distributions and refractive indices yields the bulk aerosol properties such as extinction efficiency, SSA, and phase function. The asymmetry parameter from the Henyey-Greenstein (HG) phase function is used to represent the aerosol relative scattering during the retrieval. Mie scattering phase function has a stronger forward scattering than the HG scattering phase function but vice versa in the side scattering angles. Retrieving above-cloud AODs at forward scattering angles from HG phase function slightly underestimate above-cloud AODs since the HG phase
function has a higher backscattering than the Mie phase function. A larger backscattering would shift the LUT to a higher reflectance ratio due to a relatively higher TOA radiance.

The SEVIRI retrieval begins with a simultaneous COD-CER bi-spectral retrieval from 0.81 µm and 1.64 µm (Nakajima and King 1990) following gamma size distributions and Mie scattering theory. The Cloud C1 scattering phase function from Garcia and Siewert (1985) is used to represent the cloud phase function. MODIS CERs are used to guide the LUT development with MODIS-SEVIRI collocated pixels. This simultaneous retrieval technique relies on the reflectance ratio of the 0.64µm (R_{0.64}) reflectance to the 0.81µm (R_{0.81}) reflectance as a function of R_{0.81} (Figure 3.2), which was developed by Jethva et al. (2013) for MODIS. Likewise, this approach has also been applied to simultaneously retrieve absorption AOT and COT using radiances from OMI (Torres et al., 2012) and POLDER (Peers et al., 2015). We used reflectance values at 0.64 µm instead of those at 0.47 µm as in Jethva et al. (2013) since the 0.64 µm is the shortest available wavelength on SEVIRI.

Spectral thresholds for smoke above clouds are identified on the spectral decision surface in Figure 3.2. Results from RT calculations show that smoke above clouds of COT = 2 mainly induces scattering effect with AOT retrieval highly sensitive to the changes in reflectance ratios. In contrast, smoke aerosol above clouds of COT ≥ 6 clearly causes absorbing effects with weak sensitivity to the changes in reflectance ratios during AOT retrieval. Both R_{0.81} and R_{0.64}/R_{0.81} decrease with an increasing AOT, indicating that attenuation of reflectance by smoke occurs at both wavelengths. The attenuation, however, is greater at the 0.64µm than at 0.81µm. These trends are consistent with the LUT in Jethva et al. (2013).
Table 3.1. Input parameters in the radiative transfer model for smoke above clouds. Aerosol properties are obtained from AERONET observations in Mongu, Zambia on 13 August 2006.

<table>
<thead>
<tr>
<th>RT INPUT PARAMETERS</th>
</tr>
</thead>
<tbody>
<tr>
<td>SZA</td>
</tr>
<tr>
<td>VZA</td>
</tr>
<tr>
<td>RAA</td>
</tr>
<tr>
<td>Cloud height</td>
</tr>
<tr>
<td>Aerosol height</td>
</tr>
<tr>
<td>Cloud effective radius</td>
</tr>
<tr>
<td>Surface reflectance</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Aerosol properties</th>
<th>Wavelength (µm)</th>
<th>Single scattering albedo</th>
<th>Asymmetry parameter</th>
<th>Aerosol optical thickness</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.44</td>
<td>0.84</td>
<td>0.64</td>
<td>0.45</td>
</tr>
<tr>
<td></td>
<td>0.67</td>
<td>0.79</td>
<td>0.52</td>
<td>0.20</td>
</tr>
<tr>
<td></td>
<td>0.86</td>
<td>0.76</td>
<td>0.46</td>
<td>0.12</td>
</tr>
<tr>
<td></td>
<td>1.02</td>
<td>0.75</td>
<td>0.45</td>
<td>0.08</td>
</tr>
</tbody>
</table>

Given the complexity of AOT and COT retrievals below COT < 6, smoke above cloud detection in this study focuses on smoke above clouds of COT ≥ 6. Spectral thresholds are based on the region in the spectral decision surface that is simultaneously less than the second order polynomial along COT = 6 (blue curve) for various above-cloud AOTs and the second order polynomial along AOT = 0 for COT ≥ 6 (black curve). Note that spectral thresholds vary depending on model input parameters. Thus, Figure 3.2 only represents an LUT smoke above cloud detection for a specific set of aerosol properties, solar geometry, and viewing geometry. The second order polynomial equations that are used for the LUT are shown in Figure 3.7.

3.3.2. SEVIRI algorithm

The goal of this study is to develop an algorithm for detecting smoke aerosols above closed-cell Sc on SEVIRI at pixel-level using spectral, textural, and temporal characteristics. The algorithm development is organized as follows. Initially, we examine the spectral signatures of absorbing aerosols above clouds from SEVIRI’s reflectance and thermal channels. A spectral
decision surface is inferred from the LUT to establish spectral thresholds. Next, we apply a
textural test to assess the heterogeneity of a pixel and its neighboring pixels in order to remove
cloud edges. When a pixel satisfies both spectral and textural tests, it undergoes a set of temporal
tests for identifying smoke aerosols above closed-cell Sc.

Figure 3.2. A look-up table showing the simultaneous AOT and COT retrievals of smoke above
clouds based on SEVIRI reflectance channels. Input parameters of aerosol and cloud properties
are outlined in Table 3.1. Arrows indicate the direction to which the AOT increases in an interval
of 0.0, 0.5, 1.0, 2.0, 3.0, 4.0, and 5.0. The black solid line connects various COTs in the absence
of aerosols (i.e., AOT = 0.0). Dashed lines connect AOTs of same COTs. The two solid lines
indicate spectral thresholds for the identification of absorbing aerosols above clouds with COT ≥
6.
Explanations on color differences are outlined with spectral signatures in Figure 3.3, which illustrate means and one standard deviations of reflectance for pristine clouds, aerosols above clouds with $1 < \text{AI} < 2$, and aerosols above clouds with $\text{AI} > 2$ from all case days (Table 3.2). The spectral signatures are based on pixels with $R_{0.64} > 0.2$ to ensure that only thick clouds are chosen for the statistics. Note that pixels of all channels in Figure 3.1a have also undergone histogram equalization in order to utilize all gray level values while attaining a quasi-uniform histogram over all reflectance values. This image enhancement technique provides a stronger distinction among features for better visualization since it gives the best representation of details at all ranges of reflectance values. As an example, clouds have relatively higher reflectance values than most features in these three solar channels, so they tend to appear brightest on an RGB composite (Figure 3.3a). When aerosols lie above clouds with $1 < \text{AI} < 2$ (Figure 3.3b), solar attenuation reduces the reflectance of both the green (VIS) and the blue (NIR) channels by over 10% but by less than 5% in the red channel. The RGB composite after the entire image undergoes histogram equalization causes absorbing aerosols above clouds to appear as light grey. For $\text{AI} > 2$ (Figure 3.3c), aerosols reduce the reflectance of both green and blue, causing the image to appear as light brown.

The smoke outbreak on 13 August 2006 that emanated from central Africa and transported smoke towards the southeast Atlantic above marine stratocumulus is chosen for the algorithm development due to the high range of AI values during A-Train overpass on that day. Figure 3.1b shows that OMI AI reaches as high as 4 in the middle of this image. However, we also perform an uncertainty analysis that includes cases where OMI AI values were less than 2.5 in this region. In this study, CALIOP and OMI serve as a reference for validating aerosol and cloud presence.
3.3.2.1. Spectral signatures

The initial stage of the algorithm is based on traditional spectral signatures from both reflectance and thermal channels. First, a pixel undergoes a temperature test at 10.8 µm ($T_{10.8}$), which separates clouds from land and ocean. $T_{10.8}$ for cloud tops tends to have a relatively lower temperature than that for ocean or land since this window wavelength detects either the earth’s surface temperature under clear-sky or the cloud-top temperature when clouds are present. A pixel is required to range between 280K and 295K in order to isolate liquid clouds from either land, ocean, mixed-phase clouds, or ice clouds.

The next test involves a reflectance analysis, which is based on the spectral decision surface from the LUT in Figure 3.2. As discussed earlier, $R_{0.64}$ and $R_{0.81}$ serve as the primary SEVIRI channels for separating pristine clouds from smoke above clouds since TOA reflectance at both wavelengths are sensitive to smoke scattering and absorption effects. Figures 3.4 and 3.5 depict measurements along the green line and the red line in Figure 3.1a, respectively. These figures include information from CALIOP, OMI, MODIS, and SEVIRI. Cloud edges between latitudes of $-23.2^\circ$ and $-22.5^\circ$ are evidenced by $r_s > 20\mu m$ and COT < 2 due to the emergence of multiple solutions for simultaneous low VIS and NIR reflection functions (Nakajima & King, 1990). As seen in Figure 3.1a, this region is dominated by broken clouds, which supports the likelihood of cloud edges during the CALIOP overpass. Moreover, this region consists of mainly pristine clouds since OMI AI is below 0.5 and the VFM does not indicate any aerosols above low-level clouds in this region. The equatorward decrease of CER and the increase of COT indicate the transition from cloud edge towards optically thick cloudy areas.
Table 3.2. Number of A-train pixels along the CALIOP track that were used for uncertainty analysis.

<table>
<thead>
<tr>
<th>CALIOP track no. (date)</th>
<th>Number of pixels</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 (13 August 2006)</td>
<td>490</td>
</tr>
<tr>
<td>1 (31 August 2006)</td>
<td>435</td>
</tr>
<tr>
<td>2 (24 August 2006)</td>
<td>446</td>
</tr>
<tr>
<td>3 (10 August 2006)</td>
<td>445</td>
</tr>
<tr>
<td>4 (18 July 2006)</td>
<td>178</td>
</tr>
<tr>
<td>5 (27 July 2006)</td>
<td>155</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>2149</strong></td>
</tr>
</tbody>
</table>

Figure 3.3. Spectral signatures of (a) pristine clouds, (b) aerosols above clouds for 1<AI<2, and (c) aerosols above clouds for AI > 2 from all case days in terms of mean and one standard deviation in each channel. The red channel (R), the green channel (G), and the blue channel (B) have central wavelengths at 1.64 µm, 0.81 µm, and 0.64 µm, respectively.
The southern tip of the green transect consists of ocean. Both $R_{0.64}$ and $R_{0.81}$ are less than 0.05 and $R_{0.64} > R_{0.81}$ south of $-23.5^\circ$ as shown in Figure 3.4. At $-23.5^\circ$, $R_{0.64}$ and $R_{0.81}$ are ~0.16 and ~0.17, respectively. This reflectance combination suggests that the retrieval would take place outside the spectral thresholds. Figure 3.5 shows a transect that consists of high aerosol absorption above clouds as indicated by $AI \geq 2$ throughout the entire transect. As an example, the reflectance ratio of ~0.865 ($R_{0.64}/R_{0.81} = 0.45/0.52$) and $R_{0.81}$ of ~0.52 at $-18.0^\circ$ on this transect corresponds to symbol “+” on the LUT in Figure 3.2, yielding an AOT and a COT value of ~1.2 and ~17, respectively.
Figure 3.4. (a) The green line in Figure 3.1a illustrating $R_{0.64}$ and $R_{0.81}$ in SEVIRI (at 1330 Z), COT and CER from MYD06 (at 1335Z), and OMI AI (at 1315 Z) grouped into four categories ($< 0.5$, $0.5 – 1.0$, $1.0 – 2.0$, and $> 2.0$) on 13 August 2006. (b) The heterogeneity metric based on a group of $3 \times 3$ pixels from SEVIRI at the 0.64 µm channel.
Figure 3.5. As in Figure 3.4, but for the red line in Figure 3.1(a).
3.3.2.2. Textural statistics

While spectral thresholds provide wavelength-dependence of reflectance and temperature for smoke above cloud identification, the textural analysis reveals spatial characteristics of a group of pixels surrounding the pixel of interest. The heterogeneity metric (HM) performs the best among several textural measures in isolating cloud centers from cloud edges according to our uncertainty analysis and visual inspection from all case days (Table 3.2). The HM of a pixel is obtained by calculating the ratio of the standard deviation of $R_{0.64}$ to the mean of $R_{0.64}$ for a group of $3 \times 3$ pixels. The HM has also been applied to distinguish cloud edges from the center of extensive thick clouds in Liang et al. (2009). Such a task is not well captured by spectral thresholds alone.

Figure 3.4b shows that the HM of the ocean is low ($\sim 2.5 \times 10^{-4}$) because of its high homogeneity. Between $-23.5^\circ$ and $-22.7^\circ$, the HM reaches as high as $\sim 1.2 \times 10^{-2}$ due to a combination of cloud edges and open-cell Sc. From $-22.7^\circ$ equatorward, the HM drops to $\sim 10^{-3}$, indicating that this region is more homogeneous than cloud edges. Figure 3.5b reveals that the HM values are on the order of $10^{-4}$, indicating that reflected surfaces are fairly homogeneous throughout this domain. Based on our visual inspection and uncertainty analysis, a typical cloud center has an HM ranging from $2 \times 10^{-4}$ to $6 \times 10^{-3}$. Values below this threshold are likely as homogeneous as ocean, while values above this threshold are likely cloud edges.

3.3.2.3. Temporal analysis

Using geostationary data allows one to assess the short-term evolution of clouds and aerosols because of their high temporal (15-min) resolution. While both spectral signatures and textural analysis are essential for detecting smoke aerosols above cloud centers, scrutinizing a pixel over a period of time offers another dimension of analysis. In this case, the consistency of
R_{0.64} over a time period provides the capability of synchronously assessing cloud coverage and cloud advection. Subsequently, one can determine pixels that have horizontally uniform cloud distributions (i.e., closed-cell Sc). For brevity, we refer to this set of tests as the “temporal consistency” tests.

The change in R_{0.64} at the four colored circles along the CALIOP overpass in Figure 3.1a is presented in Figure 3.6. The location of the four colored circles are selected based on similar MYD06 COTs with increasing AI. SEVIRI at 1330 Z is collocated with A-Train at nearest time and space. The white, black, orange, and blue circles are located at OMI AI values of 0, 1, 2, and 3.5, respectively. In the AI = 0 case (white circle), CALIOP only detects the presence of a cloud layer, with COT = 15 according to MYD06. At the collocated pixel, SEVIRI detects R_{0.64} of 52 %. In the preceding times (i.e., T – 30 and T – 15) at the identical pixel location, R_{0.64} were 46 % and 61 % at 1300 Z and 1315 Z, respectively. It can be seen that R_{0.64} at this pixel location was rather unstable in terms of the change in magnitude. Moreover, the alternating change in the sign of R_{0.64} in each time interval clearly suggests that this region consists of a combination of broken clouds or cloud edges. In the succeeding times (i.e., T + 15 and T + 30), R_{0.64} dropped to 37 % and 26 %, respectively. While these visible reflectance values still warrant the presence of clouds, the cloud distribution over this region is non-uniform. Hence, this pixel unlikely represent a closed-cell Sc. Figure 3.1a confirms that cumulus clouds dominate the white circled region.
Figure 3.6. Temporal variations of $R_{0.64}$ (in %) for the four colored circles along the CALIOP overpass in Figure 3.1a during five consecutive SEVIRI snapshots with 15-minute interval. Values in each small rectangle represents a $R_{0.64}$ of a SEVIRI pixel. The four black-shaded rectangles at 1330Z represent the SEVIRI pixels that are collocated with OMI AI, CALIOP, and MYD06 in both time and space. The boldfaced numbers at the center of each $3 \times 3$ pixels outside 1330Z are collocated with A-Train overpass in location but not in time.

The other three circles show examples of positive AI values above clouds. In the case of AI = 1.0 (black circle), $R_{0.64}$ at the collocated pixel was 55% at 1330 Z. In the preceding times, $R_{0.64}$ increased from 41% at 1300 Z to 48% at 1315Z. However, the reflectance plummeted to 42 % at 1345 Z and then dropped down to 28% by 1400Z, indicating that this pixel was under broken cloud coverage. For AI = 2.0 (orange circle), $R_{0.64}$ ranged between 47 % and 54 % at the collocated pixel during the five time frames. For AI = 3.5 (blue circle), $R_{0.64}$ ranged between 39 % and 46 % at the collocated pixel during the five time frames. Both the orange and the blue circles are located at an overcast closed-cell Sc region as shown in Figure 3.1a. Our findings suggest that closed-cell Sc is well represented when $R_{0.64}$ changes by less than 25 % in every 15-min interval during the four time intervals referencing from the time of higher $R_{0.64}$ between each time interval. As an example, pixels over both the orange and the blue circles satisfy the 25 %
criteria in all four time intervals. For the white circle, however, $R_{0.64}$ change exceeded 25% between 1330 Z – 1345 Z and between 1345 Z – 1400 Z even though the two preceding time intervals changed by less than 25%. In this case, the white circle area would not be assigned as closed-cell Sc based on the temporal consistency thresholds.

Figure 3.7 illustrates a series of tests that each SEVIRI pixel must undergo before being assigned as a smoke above closed-cell Sc pixel. A pixel will follow the solid arrow if it satisfies the criteria in a particular test; otherwise, it will follow the dashed line. For a pixel to be qualified as smoke above closed-cell Sc, it must pass the spectral, textural, and temporal consistency tests. Italic texts within each dashed box indicate features that a pixel may represent if a test is not satisfied. All thresholds have undergone iterative adjustments based on the uncertainty analysis and visual inspection over the five case days.

Figure 3.7. An algorithm for detecting smoke above clouds using spectral signatures, textural analysis, and temporal consistency tests based on SEVIRI. Solid boxes are tests that each pixel must undergo starting from the top. Boxes in white, grey, and black are spectral signature tests, the textural test, and the temporal consistency tests, respectively. When a pixel does not satisfy the criteria in a particular test, it will follow the dashed arrow, which shows the feature that it likely represents.
3.3.2.4. Algorithm results

Results after applying the smoke above closed-cell Sc algorithm are shown in Figure 3.8. The original RGB image prior to algorithm application is shown in Figure 3.8a for readers’ reference, which is identical to Figure 3.1a after removing all the labels. Results from applying only the spectral thresholds and textural statistics of the algorithm are presented in Figure 3.8b. The central portion of this image is classified as smoke above clouds, where OMI AI values exceeds 0.5. However, smoke above clouds are assigned by the algorithm in the region bounded by 18 – 20°S and 0 – 4°W where OMI AI values are below 0.5. In the southwestern corner of the image, a scattered amount of pixels is assigned as smoke above clouds, coinciding with a few AI = 0.5 contours. Figure 3.8c shows results after incorporating the temporal consistency tests into the algorithm. Classification criteria become more stringent than spectral and textural tests alone since the temporal consistency tests determine if the pixel of interest is closed-cell Sc. An examination of the synoptic weather map indicates that clouds in the southwestern portion of the image are likely associated with a cold front. Very few pixels satisfy the criteria for smoke above closed-cell Sc in this area after applying the temporal consistency tests, affirming the soundness of applying the entire series of algorithm for smoke above closed-cell Sc detection.

3.4. Uncertainty analysis

The uncertainty analysis is conducted using CALIOP cloud and aerosol layer product, the VFM, and OMI AI as the reference data for comparison with SEVIRI-derived smoke above cloud algorithm. The criteria for smoke above clouds detection by CALIOP include the criteria discussed in Section 3.3 and OMI AI exceeding 0.5. For the pristine cloud detection case, the criteria of reference data include CALIOP layers with clouds below 5km, aerosol-free, OMI AI below 0.5, and SEVIRI R0.64 exceeding 0.2. The inclusion of SEVIRI in the reference data is
required to ensure that both CALIOP cloud layers and a SEVIRI pixel agree on the cloud presence.

Figure 3.8. (a) The RGB image as in Figure 3.1a. (b) Results of SEVIRI-derived smoke above clouds based on only spectral and textural tests. (c) Results of SEVIRI-derived smoke above closed-cell Sc based on spectral, textural, and temporal consistency tests. Red contours denote lines of equal OMI AI values starting from 0.5. The honeycomb-like contours along the coastlines results from row anomaly. Note that OMI AI contours in both (b) and (c) are identical.

The uncertainty analysis is presented in terms of an user accuracy and a producer accuracy. The user accuracy is defined as the percentage of SEVIRI pixels that are correctly identified as smoke above any cloud type rather than as pristine clouds when compared with the reference data. The producer accuracy is the percentage of reference data pixels that detects smoke above clouds and is also agreed by SEVIRI. Note that since the cloud reference data (i.e., CALIOP) do not provide information on cloud types (i.e., open-cell or closed-cell), the uncertainty analysis is limited to smoke above clouds rather than smoke above closed-cell Sc. However, this study compares the accuracy between the inclusion and the exclusion of the temporal consistency tests to assess the impact of temporal information on the resulting accuracy.
The bar graph in Figure 3.9 shows the user accuracy and the producer accuracy converted from a traditional error matrix (or confusion matrix). Using only the spectral signature and textural tests, the SEVIRI algorithm has identified 219 smoke above clouds pixels with 108 pixels being verified by the reference data. Thus, the user accuracy for this scenario is ~49% (“S+T” in Figure 3.9). When adding the temporal consistency tests in the SEVIRI algorithm, a total of 162 SEVIRI pixels have been identified as smoke above clouds. Among these 147 pixels, 96 pixels are being verified by the reference data, yielding an user accuracy of ~65% (see “ALL” in Figure 3.9). Hence, the increasing user accuracy after incorporating the temporal consistency tests suggests that the algorithm performs better at detecting smoke above closed-cell Sc than smoke above mixture cloud types.

As aforementioned, 96 pixels have been confirmed as smoke above clouds by both the reference data and the algorithm when the temporal consistency tests were included. In 28 collocated pixels, the reference data indicates smoke above clouds while the SEVIRI algorithm detects either pristine clouds or smoke above thin clouds. As a result, the producer accuracy is ~77% (see “PROD” in Figure 3.9). Likewise, the producer accuracy when excluding the temporal consistency tests (spectral and textural only) is ~69% (not shown). In summary, the SEVIRI algorithm generally identifies smoke above clouds when CALIOP also identifies the same feature at the collocated pixel. The producer accuracy exceeds the user accuracy regardless of the inclusion of the temporal consistency tests since CALIOP has the tendency to underestimate the presence of thin smoke aerosols above liquid clouds during daytime (Liu et al., 2015; Torres et al., 2013). We focus on the producer accuracy in this study since we aim to identify smoke above clouds provided that CALIOP also detect the same feature in the collocated region. Additionally, the temporal consistency tests also improve the accuracy
because closed-cell Sc is characterized with more homogeneous background than cloud edges or cumulus clouds.

Figure 3.9. Bar graph depicting the accuracies of SEVIRI-derived smoke above clouds. The “S + T” label denote the user accuracy using only a combination of spectral and textural tests. The “ALL” label denotes the user accuracy for all (spectral, textural, and temporal consistency) tests. The “PROD” label denotes the producer accuracy for all tests.

3.5. Summary

An algorithm for detecting smoke above closed-cell Sc has been developed for the SEVIRI data set, which relies on spectral signatures, textural statistics, and high temporal resolution capabilities. The southeast Atlantic is dominated by high biomass burning smoke aerosol loadings and semi-permanent Sc during Austral winters, serving as a natural laboratory for developing the smoke above closed-cell Sc algorithm. The algorithm is generated by collocating the SEVIRI data with A-Train satellite sensors including MODIS, CALIOP and
OMI. CALIOP provides the vertical distributions of aerosols and clouds while the OMI AI provides UV aerosol absorption above clouds.

The algorithm comprises the use of spectral signatures from both reflectance and thermal channels with prescribed thresholds. A spectral decision surface inferred from an LUT is generated to assign spectral thresholds for identifying smoke above clouds. This LUT relies on reflectance ratio of $R_{0.64}$ to $R_{0.81}$ and provides the capability of synchronously retrieving AOT and COT. Importantly, this algorithm mainly detects smoke above clouds of $COT \geq 6$ since identifying smoke above thin clouds is subject to high uncertainty. The HM, a textural analysis used in this study, distinguishes cloud edges from cloud center. Upon passing the spectral and textural tests, a pixel undergoes a set of temporal consistency tests, which inspects pixel consistency and homogeneity over a 60-min period in $R_{0.64}$.

The uncertainty analysis is based on the user accuracy and the producer accuracy from the five selected case days in 2006. The user accuracy is defined as the percentage of SEVIRI pixels that are correctly identified as smoke above clouds rather than as pristine clouds with respect to A-Train. The producer accuracy is defined as the percentage of reference pixels that are identified as smoke above clouds and is simultaneously agreed by SEVIRI. Results indicate that the user accuracy is $\sim 49\%$ when only applying the spectral and textural tests and increases to $\sim 65\%$ when incorporating the temporal consistency tests. The producer accuracy of the algorithm in this study is $\sim 69\%$ ($\sim 77\%$) when excluding (including) the temporal consistency tests, implying that the SEVIRI algorithm generally identifies smoke above clouds when CALIOP also identifies the same feature at the collocated pixel. However, when a SEVIRI pixel is classified as smoke above cloud, CALIOP only agrees with the collocated SEVIRI pixel $\sim 49\%$ of the time. These findings are anticipated since CALIOP has the tendency to underestimate the presence of
thin smoke aerosols above liquid clouds during daytime (Liu et al., 2015; Torres et al., 2013).
The accuracy in the present study focuses on the producer accuracy since our goal is to validate
the presence of smoke above clouds from SEVIRI provided that CALIOP also identifies the
same feature at the collocated pixel.
CHAPTER FOUR

SATELLITE RETRIEVAL AND AIRCRAFT MEASUREMENTS OF ABOVE-CLOUD AEROSOL OPTICAL PROPERTIES

4.1. Overview

Passive satellite measurements provide a global coverage of aerosols, which are important for calculating the aerosol DREs. However, the operational aerosol products are presently only available over cloud-free regions, which exclude aerosols above clouds. The MODIS aerosol operational product (Levy et al., 2013), for example, currently only provides aerosol information over cloud-free regions and neglects areas of above-cloud aerosols. Therefore, estimating aerosol DREs using only the standard product does not yield an accurate estimate.

It is imperative to correctly determine the aerosol and background properties when evaluating the aerosol DRE. The aerosol DRE in cloud-free regions over oceans generally leads to cooling at TOA and reduces surface temperatures since aerosols scatter more solar radiation than that of the ocean surface. The land surface types affect both the magnitude and the sign of the estimated aerosol DRE. For example, the aerosol DRE over sandy surfaces is $-1.54 \text{ W m}^{-2}$ but changes to $+1.69 \text{ W m}^{-2}$ over snow surfaces over the Himalayas (Nair et al., 2013).

The foremost above-cloud aerosol analysis starts with identifying the location of their
presence. In multi-spectral satellite remote sensing, the identification of above-cloud biomass burning aerosols begins with a spectral analysis using bi-spectral color-ratio techniques (Chang & Christopher, 2016; Jethva et al., 2013) or spectral optimization techniques (Meyer et al., 2015; Sayer et al., 2016). Since the knowledge of AOTs and COTs are required for estimating the above-cloud aerosol DREs, both parameters must be retrieved to the best ability using RT theory. COTs are functions of visible reflectance (Greenwald & Christopher, 2000; Nakajima & King, 1990) but are negatively biased in the presence of overlying absorbing aerosols (Chang & Christopher, 2016; Haywood et al., 2004; Jethva et al., 2013). Therefore, a COT correction must be performed while retrieving AOTs prior to estimating the DREs. Such a task necessitates a simultaneous retrieval of above-cloud AODs and underlying COTs from multi-spectral sensors using spectral techniques. However, above-cloud Aerosols above thin clouds (typically COT < 4) are usually neglected given the large uncertainties at distinguishing AOT and COT in these circumstances. Likewise, retrieving large aerosol loadings above optically thick clouds is also challenging since the attenuated radiances of cloud scattering by heavy aerosol loading could have the identical radiance as low AOT above low COT. Errors in optical depth retrieval would bias the estimate of DREs on top of SSA errors (Feng & Christopher, 2015; Meyer et al., 2015).

Above-cloud aerosol retrievals are not limited to multi-spectral sensors in satellite remote sensing. Satellite retrieval approaches from polarization measurements also provide the capability for retrieving above-cloud AOTs (Waquet et al., 2009). A novel strategy for retrieving above-cloud fine mode AOTs using polarized phase function at forward scattering angles from the POLDER has been developed (Waquet et al., 2013). Such an approach has also been applied to simultaneously retrieve the aerosol absorption optical thickness and the underlying COT from POLDER measurements (Peers et al., 2015). Simultaneous retrieval of AOT and COT has been
conducted using inversion methods from near-UV radiances in OMI (Torres et al., 2012) and from visible to near-infrared color-ratio techniques in MODIS (Jethva et al., 2013). Chang and Christopher (2016) have also applied the color-ratio technique to identify above-cloud aerosols from SEVIRI, which can also be used for retrieval purposes. Meyer et al. (2015) have developed an optimal estimation technique to simultaneously retrieve above-cloud AOT and underlying liquid cloud optical and microphysical properties by utilizing six MODIS channels ranging from VIS to shortwave infrared wavelengths.

Spaceborne satellite sensors continuously acquire measurements, rendering large data quantities for constructing climatological data sets and enabling the computation of robust climatological results. Passive satellite sensors with large fields-of-view (e.g., MODIS, and SEVIRI) measure TOA radiances and require inversion algorithms to retrieve various aerosol and cloud parameters that are necessary to calculate DREs. However, TOA radiance inversions are accompanied by numerous biases including atmospheric gaseous absorptions, Rayleigh scattering attributed to aerosol-cloud height assumptions, land surface complexities, assumptions on aerosol intrinsic properties (i.e., SSA and phase function), and cloud particle size distribution assumptions (Meyer et al., 2015). Thus, in situ observations from aircraft field campaigns are essential to evaluate errors and uncertainties in satellite observations. The ongoing ORACLES field experiment aims to investigate aerosol-cloud-radiation interactions over the southeast Atlantic, creating an excellent opportunity for validating above-cloud AODs and underlying CODs from satellite retrievals. Prior to the ORACLES field experiment, three field experiments have examined aerosol transport and chemical compositions in the south Atlantic, namely Transport and Atmospheric Chemistry Near the Equator-Atlantic (TRACE A) (Bachmeier & Fuelberg, 1996), SAFARI-1992 (Swap et al., 1996), and SAFARI-2000 (Swap et al., 2003).
During SAFARI-2000, the C-130 aircraft conducted flights off coasts of Namibia and Angola. It was reported that the top and the base of biomass burning aerosols are located at about $5120 \pm 550$ m and $1520 \pm 660$ m, respectively (Haywood et al., 2004). During the flight on 7 September 2000, biomass burning aerosols were found to be located at altitudes between 1.8 – 3.7 km, whereas clouds are located below 1 km (Keil & Haywood, 2003).

A few comparisons between satellite measurements, ground-based observations and field experiments have been conducted over west Africa. During Dust and Biomass-burning Aerosol Experiment (DABEX) (Johnson et al., 2008), dust and biomass burning aerosol properties have been compared among UK Facility for Airborne Atmospheric Measurements (FAAM), AERONET, and satellite measurements in west Africa (Johnson et al., 2009). It was found that biomass-burning AODs between in situ and the Banizoumbou AERONET observations were within about 0.05 from each other. The MODIS deep-blue and MISR AODs agreed with AERONET observations to within 10%. Saharan desert dust AODs and aerosol vertical distributions from satellite measurements have been compared with UK FAAM during Geostationary Earth Radiation Budget Intercomparisons of Longwave and Shortwave (GERBILS) (Christopher et al., 2009, 2011; Haywood et al., 2011). Overall, it was noted that satellite AOD retrievals and in situ AOD values agreed to within 0.1 – 0.2 for low AOD (AOD < 1). However, satellite products generally underestimates AODs in regions where in situ AODs exceed 1.

Above-cloud aerosols exist in numerous parts of the world’s oceans such as southeast Atlantic, tropical northeast Atlantic, northwest Pacific, southeast Pacific, The Arabian Sea, and Gulf of Tonkin (Devasthale & Thomas, 2011; Zhang et al., 2016). Nonetheless, the southeast Atlantic has the highest frequency of above-cloud biomass burning aerosols during Austral
winter. An inter-comparison of above-cloud AODs have been performed among A-Train sensors (Jethva et al., 2014), but a comparison of above-cloud AODs between satellite and aircraft data has not been performed. Above-cloud AODs from satellite can only be validated by measuring direct solar transmittance above cloud. This goal can only be achieved from aircraft measurements rather than AERONET observations. Thus, the aircraft field campaign such as ORACLES dedicated to studying above-cloud biomass burning aerosols serve as a crucial step to evaluate satellite retrieval performance of above-cloud aerosols in the southeast Atlantic. The ORACLES field campaign also concurrently collaborates with other field experiments to consolidate the in situ measurements over the southeast Atlantic (Zuidema et al., 2016).

The primary goal of this study is to validate retrievals of above-cloud aerosol and underlying cloud properties from MODIS and SEVIRI against 4TSAR on board the P3 aircraft during four case days of ORACLES 2016. Section 4.2 describes the 4STAR instrument and satellite data sets. Section 4.3 outlines the area of study. Methods for satellite retrievals and data filtering are explained in Section 4.4. Results and summary are presented in Sections 4.5 and 4.6, respectively.

4.2. Data

4.2.1. 4STAR instrument

The 4STAR sun photometer onboard NASA ORACLES P-3 aircraft, a successor to NASA Ames airborne tracking Sun photometers (AATS), serves as a benchmark to compare above-cloud AODs with satellite-retrieved above-cloud AODs (Dunagan et al., 2013; Shinozuka et al., 2013). At a 1-Hz measurement frequency, 4STAR measures hyperspectral `columnar AOD between 350-1700 nm at about 1 nm spectral resolution based on the direct solar beam transmittance. The direct transmittance are measured exterior to the aircraft using a tracking head
that locks normal to the direct solar beam. The tracking head is structured by a quadrant
differential photodiode sensor that controls the azimuth and elevation motors. This study uses the
calibrated 4STAR AOD product with correction accounting for window deposition that builds up
during the flight. The 4STAR instrument has been used to examine pollution-induced trace gases
in northeast US (Segal-Rosenheimer et al., 2014) and evaluate hyperspectral AODs against
AERONET measurements (Shinozuka et al., 2013) during TCAP over northeast US. Figure 4.1
shows means and standard deviations of AOD spectral dependence from 4STAR for ORACLES
2016, which took place in September 2016. The spectral pattern indicates fine-mode particles
with a mean Angstrom Exponent (470 and 865 nm) of about 1.78. AOD standard deviations are
relatively higher at shorter wavelengths since extinction coefficients of biomass burning aerosols
are higher at shorter wavelengths, which is a typical characteristic of biomass burning aerosols
(Bergstrom et al., 2007). Stronger Rayleigh scattering and gaseous absorption at shorter
wavelengths also interferes with AOD estimates from 4STAR.

Figure 4.1. 4STAR means (dots) and standard deviations (error bars) of spectral AODs during
ORACLES 2016. An exponential fitting of AOD means is shown with a black line.
4.2.2. Satellite data

MODIS is a multi-spectral instrument consisting of 36 spectral (0.405 µm - 14.385 µm) channels with a swath width of ~2330 km with a near-global daily coverage. The 1-km calibrated radiances (MYD021KM) from the solar channels are used to simultaneously retrieve pixel-level above-cloud AODs and underlying cloud properties over the ocean. The MODIS cloud mask product (MYD35_L2) is used to ensure that above-cloud AOD retrievals in this study are performed only over the ocean. Presently, MODIS aboard both the Terra and the Aqua satellites but only Aqua participates in the A-Train constellation. Aqua overpassed the same region as the ORACLES afternoon flight during ORACLES 2016.

Additionally, SEVIRI on board MSG consists of eleven spectral channels (three solar channels, and eight thermal channels) and a high resolution visible (HRV) channel covering half of the full disk in the E-W direction and a full disk in the N-S direction over Europe, Africa, and the Atlantic Ocean (Schmetz et al., 2002). With a 15-minute repeat cycle and a 3-km nadir spatial resolution, this data set is applicable to assessing the diurnal cycles of various parameters. As in Section 3, the 0.64 µm and the 0.81 µm are used to simultaneously retrieve above-cloud AODs and underlying CODs.

4.3. Study area

The southeast Atlantic is dominated by high biomass burning smoke aerosol loadings and semi-permanent stratocumulus clouds during Austral winters, serving as a natural laboratory for examining above-cloud aerosol optical properties (Devasthale and Thomas, 2011; Zhang et al., 2014, 2016). Aircraft data from four days during the ORACLE 2016 field experiments are selected for validating above-cloud AODs against MODIS and SEVIRI. These cases are chosen as the suitable days for comparison by both aircraft and satellite measurements. The filtering
criteria for both types of measurements are discussed in Section 4.4. Figure 4.2 shows a MODIS true-color image overlaid with a portion of ORACLES flight track that are plotted by AOT values. Note that the science flights are primarily conducted mainly in the northern portion of each day. However, measurements taken throughout the transect are included in the validation if both satellite and aircraft data satisfies the filtering criteria.

Figure 4.2. Aqua-MODIS true-color images on (a) 2nd, (b) 6th, (c) 14th, and (d) 20th September 2016 over the southeast Atlantic. A portion of ORACLES P3 flight track that include 4STAR AOT values are shown for each day.
4.4. Methods

The MODIS above-cloud AOD retrieval begins with a simultaneous COD and CER retrieval using the 1.24 µm and 2.13 µm pair. However, only the retrieved CER is needed for the next step, which involves a simultaneous above-cloud AOD and underlying COD retrieval if the COD > 4 in a pixel. Uncertainties of COD retrieval below aerosol layer become exceedingly high when COD < 4, so simultaneous AOD and COD retrieval is excluded (Meyer et al., 2015). When CODs exceed 4, CERs are then used to simultaneously retrieve AODs and CODs in the LUT based on the color-ratio technique from 0.470 µm and 0.865 µm channel (Jethva et al., 2013) assuming aerosol properties in Mongu, Zambia as outlined in Table 3.1. The stronger solar absorption of smoke in the shorter wavelength provides the premise for the retrieval due to wavelength dependence of absorption coefficient (Meyer et al., 2015). The simultaneous AOD and COD retrieval aims to correct the low-biased COD (hereinafter called “corrected CODs”).

SEVIRI’s above-cloud AOD and underlying COD retrieval strategy follows the LUT on Figure 2.1, which is based on the color-ratio of 0.635 µm and 0.810 µm. As with MODIS, simultaneous above-cloud AOD and underlying COD retrievals are proceeded when COD > 4. Similar to Chang and Christopher (2016), the heterogeneity metric, defined as the ratio of standard deviation to the mean among a group of pixels, is used to filter pixels that are at cloud edges or are heterogeneous on both sensors.

An examination of AOD transect among 4STAR, MODIS, and SEVIRI are done along the afternoon flight path in September 2016. Only 4STAR AODs that exceeds 0.05 with uncertainties less than 0.5 are selected for comparison against satellite. The raw 4STAR AODs are available at every second and are averaged to 60 seconds for validation. Only measurements taken between 500 – 1600 m flight altitude are considered so that most of the columnar AOD can
be obtained. Below-aircraft AODs so that underestimated AODs are excluded from comparison. MODIS and SEVIRI have a spatial resolution of about 0.01° and 0.03°, respectively, along the transect of interest, so the number of data points along the transect for MODIS exceed that for SEVIRI.

4.5. Results

Figure 4.3 shows the MODIS-retrieved above-cloud AODs for 20 September 2016, which is the same day as in Figure 4.2d. This day is characterized with high aerosol loading above optically thick clouds. Above-cloud AODs increase in the northwestward direction within this area with a mean and standard deviation of 0.37 and 0.13, respectively. COD distributions resemble cloud patterns in Figure 4.2d, which are relatively higher in the eastern portion than the western portion. The mean and standard deviation of COD in this area is 11.5 and 3.7, respectively. Figure 4.4 shows the retrievals from SEVIRI for the same region as Figure 4.3, which indicates AOD with a mean and standard deviation of 0.50 and 0.11. SEVIRI’s underlying CODs show a mean and standard deviation of 12.2 and 3.4 in this domain, respectively. These findings suggest that AODs and CODs compensate each other during the retrieval since both the mean CODs and AODs in SEVIRI are relatively higher than those in the MODIS retrieval.

A comparison among 4STAR AODs with both MODIS and SEVIRI for 20 September 2016 is shown in Figure 4.5. 4STAR AODs at 550 nm are relatively constant, ranging between 0.46 and 0.49 along the entire transect, indicating that this long-range aerosol plume is relatively uniform over the region. Overall, the MODIS and SEVIRI above-cloud AOD shows a agreement with 4STAR measurements to within 0.1 assuming a SSA of 0.84 at 0.47 µm and 0.79 at 0.865 µm (Figure 4.5a). The MODIS mean AOD along the transect is ~0.43 whereas the SEVIRI mean
AOD is ~0.56. The overestimated SEVIRI above-cloud AOD suggest the existing retrieval error due to SEVIRI’s spectral limitations.

Both MODIS- and SEVIRI-retrieved underlying CODs agree with each other to within one retrieved COD interval (Figure 4.5b). MODIS CODs are generally equal or greater than SEVIRI CODs by 2. The relationships between above-cloud AODs and underlying CODs are apparent in both MODIS and SEVIRI, where relatively higher above-cloud AODs are compensated by a relatively higher underlying CODs. HM (Figure 4.5c) along the transect shows a wavy pattern that reveals cloud centers and cloud edges of the stratocumulus clouds. However, the magnitudes of the heterogeneity is too low to declare a significant retrieval bias attributed to pixel heterogeneity.

Figure 4.3. (a) Retrieved above-cloud AODs and (b) adjusted underlying CODs from Aqua on 20 September 2016. The vertical line denotes the ORACLES flight path for that afternoon.
Figure 4.4. Same as Figure 4.3, but for SEVIRI.
Figure 4.5. AOD, COD, reflectance, and heterogeneity metric from MODIS and SEVIRI along the flight path shown in Figure 4.3 on 20 September 2016. Dots are 4STAR, triangles are MODIS, and squares are SEVIRI.
The assumed SSA affects the retrieved AOD more significantly than the measurement biases. Figure 4.6 shows the results of AODs and CODs along the same transect with a weaker absorption (i.e., higher SSA) than those in Figure 4.5. The retrieved AODs (Figure 4.6a) are based on the Namibe, Angola AERONET observations, which is near the coastline of Angola away from the fire burning source. Retrieved AODs from both satellites are clearly higher than the case that assumes a stronger aerosol absorption, which is consistent with retrieval relationship between AOD and SSA for aerosols above optically thick clouds (Jethva et al. 2013, Meyer et al. 2015). At the time of this writing, SSA retrievals from ORACLES are still at preliminary stage. The calibrated SSA from in situ observations will establish an important step for parameterizing the assumed single scattering properties in RT calculations for retrieving above-cloud AODs. Interestingly, coastal SSAs do not necessarily represent a more accurate aerosol properties in the southeast Atlantic than those near the fire source. Hence, SSAs vary significantly from case-to-case. For example, Haywood et al. (2003) noted a mean SSA of 0.91 at 550 nm for aged regional haze during the SAFARI-2000 field experiment, which is more scattering than Namibe, Angola aerosol properties. Regional haze may not necessarily have more scattering properties than the fresh plume because of the presence of coated organic compounds that can enhance absorption, especially in the UV and short visible wavelengths. Organic carbons are found to be sensitive to the imaginary refractive index in addition to black carbons for internally mixed aerosols (Xie et al., 2017). Organic carbons can also enhance aerosol absorption via “lensing” effect by refocusing solar beam towards the particle center where black carbon is concentrated (Lack et al. 2012; Saleh et al. 2015). These impacts complicate the estimations of SSA, which, in turn, influence the estimates of aerosol DRE.
Figure 4.6. AOD retrievals along the same transect as in Figure 4.5 but using Namibe, Angola AERONET aerosol properties.

Figure 4.7 compares the above-cloud AOD statistics among the four days during 2016 campaign where satellite retrievals are performed. 4STAR has the lowest AOD standard deviations among the three data sets since measurements are based on direct solar transmittance. Any significant standard deviations within a tens of kilometers would be attributed to anomalous smoke emission. Since the SEVIRI color-ratio retrieval is performed at 0.635 µm instead of 0.470 µm as in the MODIS retrieval, the columnar AOD and COD would be expected to generate more uncertainties than a MODIS retrieval. The standard deviations of above-cloud AODs from SEVIRI in all days have a range between 0.16 and 0.36. However, the standard deviations of above-cloud AODs from MODIS range between 0.04 and 0.80 among the four days. These findings are consistent with the relatively more homogeneous radiances in SEVIRI compared to MODIS radiances due to SEVIRI’s coarser resolution.

The mean above-cloud AODs among the three data sets agree to within 0.07 on 2 September 2016, but the standard deviation of SEVIRI retrievals are the highest among these observations. Above-cloud aerosols are below 0.2 in the afternoon flight on 14 September 2016, leading to a challenging retrieval from satellite. Mean AODs from MODIS and SEVIRI are 0.68 and 0.53, respectively. CODs on this day are also characterized with values below 5, which barely satisfies the COD retrieval criteria. The LUT also indicates the high sensitivity of AOD to
the color-ratio at low CODs, which cause a large bias on that day. The mean above-cloud AODs between MODIS and 4STAR are only differ by 0.03 on 20 September 2016. The day is accompanied by AOD of ~ 0.5 above COD between 8 – 12, which are less sensitive to color-ratio changes of AODs than for aerosols above thin clouds.

Figure 4.7. Means and standard deviations of above-cloud AODs for 4STAR (black), SEVIRI (red), and MODIS (blue) during ORACLES 2016 field campaign for selected days. The number of samples for a particular day is denoted by “N.”
4.6. Summary

Validation of 4STAR above-cloud AODs on board P3 aircraft during ORACLES 2016 are conducted against MODIS and SEVIRI. Above-cloud AOD retrieval generally agree to within 0.1 above homogeneous clouds with COD > 8. However, above-cloud AOD retrieval are biased high when retrieving above thin clouds since the radiance separation between aerosols and clouds becomes complex. Overall, the assumed SSA results in AOD retrieval that matches well with 4STAR AOD. However, when the assumed SSA of 0.90 at 470 nm and 0.89 at 860 nm is used, the MODIS AOD are overestimated with respect to 4STAR. Satellite-retrieved AODs are clearly higher than the case that assumes a stronger aerosol absorption, which is consistent with retrieval relationship between AOD and SSA for aerosols above optically thick clouds (Jethva et al., 2013). At the time of this writing, SSA retrievals from ORACLES are still at preliminary stage. The calibrated SSA from in situ observations will set an important step to parameterize the assumed single scattering properties in RT calculations for retrieving above-cloud AODs. Thus, the present study assumes SSA from the Mongu, Zambia AERONET observation.
CHAPTER FIVE

CONCLUSIONS AND FUTURE WORK

5.1. Conclusions

The impact of seasonalities on DREs and RHRs of absorbing aerosols above clouds in the Southeast Atlantic are examined in this study. We use the SAFARI 2000 aerosol optical properties derived by Meyer et al. (2015) to build the aerosol model specifying the aerosol properties in SBDART. We also employ the aerosol optical properties from the ground-based remote sensing retrievals from the AERONET site at Mongu, Zambia to assess the impact of aerosol optical properties on DREs and RHRs. Fixed aerosol and cloud properties are used to understand implications of SZAs on the DRE and the RHR. For AOT = 0.6 at 0.55 µm located between 0 – 4 km with COT = 9.0 and CER = 12.8 µm located between 1 – 2 km under a tropical atmosphere, the diurnally averaged RHR at noon in the cloud layer is ~6.6 K day⁻¹ in June. The RHR increases to ~8.9 K day⁻¹ in October at noon due to the smaller mean SZA during this month. In June (October), the RHR in the cloud layer at noon is 1.3 (1.7) K day⁻¹ higher than the case of pristine clouds. Thus, clouds embedded in an aerosol layer experience an elevated RHR in the cloud layer compared to clouds in an aerosol-free scenario. However, the elevated aerosol layer above clouds (without aerosols in the cloud layer) reduces the RHR by ~0.2 K day⁻¹ in the cloud layer relative to a pristine cloudy case, which possibly weaken the dissipation of
cloud during daytime from a radiative standpoint. RHRs also depend on the selection of aerosol optical properties.

When all aerosols are located above liquid clouds, the DRE at TOA reaches its peak when $SZA = 54^\circ$. The DRE at TOA increases with $SZA$ from $0^\circ$ to $54^\circ$ and then decreases until $SZA = 90^\circ$, which potentially yields the same DRE for different $SZAs$ when $SZA < 76^\circ$. At 5.0°S 5.0°E, for example, the primary peak DRE of $\sim 29.5$ W m$^{-2}$ occurs at 8 Z while the secondary peak of $\sim 29.4$ W m$^{-2}$ takes place at 15 Z. At noon, the DRE at TOA are $\sim 18.9$ W m$^{-2}$, $\sim 20.5$ W m$^{-2}$, and $\sim 23.1$ W m$^{-2}$ at 5.0°S, 15.0°S, and 25.0°S along 5.0°E, respectively. The increasing DRE with latitude is expected since the DRE increases with $SZA$ when $SZA < 54^\circ$. The surface DRE responds greater to changes of $SZA$ since it increases from $-71$ W m$^{-2}$ to 0 W m$^{-2}$ when $SZA$ increases from $0^\circ$ to $90^\circ$.

An algorithm for detecting smoke above closed-cell Sc has been developed for the SEVIRI data set, which relies on spectral signatures, textural statistics, and high temporal resolution capabilities. The southeast Atlantic is dominated by high biomass burning smoke aerosol loadings and semi-permanent Sc during Austral winters, serving as a natural laboratory for developing the smoke above closed-cell Sc algorithm. The algorithm is generated by collocating the SEVIRI data with A-Train satellite sensors including MODIS, CALIOP and OMI. CALIOP provides the vertical distributions of aerosols and clouds while the OMI AI provides UV aerosol absorption above clouds.

The algorithm comprises the use of spectral signatures from both reflectance and thermal channels with prescribed thresholds. A spectral decision surface inferred from an LUT is generated to assign spectral thresholds for identifying smoke above clouds. This LUT relies on reflectance ratio of $R_{0.64}$ to $R_{0.81}$ and provides the capability of simultaneously retrieving AOT.
and COT. Importantly, this algorithm mainly detects smoke above clouds of COT $\geq 6$ since identifying smoke above thin clouds is subject to high uncertainty. The HM, a textural analysis used in this study, distinguishes cloud edges from cloud center. Upon passing the spectral and textural tests, a pixel undergoes a set of temporal consistency tests, which inspects pixel consistency and homogeneity over a 60-min period in $R_{0.64}$.

Results indicate that the user accuracy is $\sim 49\%$ when only applying the spectral and textural tests and increases to $\sim 65\%$ when incorporating the temporal consistency tests. The producer accuracy of the algorithm in this study is $\sim 69\%$ ($\sim 77\%$) when excluding (including) the temporal consistency tests, implying that the SEVIRI algorithm generally identifies smoke above clouds when CALIOP also identifies the same feature at the collocated pixel.

The assumed SSA affects the retrieved AODs and the comparisons with 4STAR AODs. Above-cloud AOD retrieval generally agree to within 0.1 above homogeneous clouds with COD $> 8$. However, above-cloud AOD retrieval are biased high when retrieving above thin clouds since the radiance separation between aerosols and clouds becomes complex. The assumed SSA of 0.84 at 470 nm and 0.76 at 860 nm results in AOD retrieval that matches well with 4STAR AOD. However, when the assumed SSA of 0.90 at 470 nm and 0.89 at 860 nm is used, the satellite AODs are overestimated with respect to 4STAR. Retrieved AODs are clearly higher than the case that assumes a stronger aerosol absorption, which is consistent with retrieval relationship between AOD and SSA for aerosols above optically thick clouds. At the time of this writing, SSA retrievals from ORACLES are still at preliminary stage. The calibrated SSA from in situ observations will set an important step to parameterize the assumed single scattering properties in RT calculations for retrieving above-cloud AODs. Thus, the present study assumes SSA from the Mongu, Zambia AERONET observation.
AR5 suggests that the radiative forcing of both aerosol-radiation interactions and aerosol-cloud interactions are $-0.45 \text{ W m}^{-2}$. The combined aerosol radiative forcing partially compensates the positive greenhouse gas forcing. This study suggests that the radiative forcing of absorbing aerosols above clouds is positive, which opposes the cloud-free aerosol radiative forcing. Accounting for the positive radiative forcing from absorbing aerosols above clouds in this study would increase the aerosol radiative forcing, resulting in a less negative aerosol radiative than that indicate in AR5. This positive forcing would cause a larger total anthropogenic radiative forcing than the current value in AR5. However, the larger total anthropogenic radiative forcing neglects cloud diurnal cycles and aerosol-cloud microphysical interactions.

5.2. Future work

The DRE of cloud-embedded absorbing aerosols and absorbing aerosols above clouds have been studied extensively using satellite observations and RT calculations. The information in this study is useful for evaluating the diversity of aerosol-cloud DRE in both satellite observations and models. This work provides data and theoretical understanding to assist positioning science flights that target measurements of AAC radiative effects. Field campaigns would provide invaluable information for improving estimates of the aerosol-cloud DRE. Scientists among several countries will collaborate and conduct various field experiments between 2016 and 2018 using aircraft and surface instruments over the Southeast Atlantic (Zuidema et al., 2016). These collaborations will offer tremendous progress in the aerosol science community.

The present study uses the Henyey-Greenstein phase function from asymmetry factor as a representation of the aerosol scattering phase function, which differs from the Mie phase
function. For purely scattering (non-absorbing) aerosols at accumulation mode, the greatest uncertainty occurs at low and high SZAs (Boucher, 1998). Magi et al. (2008) noted that using the Henyey-Greenstein phase function would approximately double the uncertainty from modeled fluxes for biomass burning aerosols. Uncertainties associated with asymmetry parameter require a detailed sensitivity analysis of aerosol microphysical properties such as particle size distributions and refractive indices. These analysis are beyond the scope of the study and should be addressed in future studies.

Furthermore, subsequent studies also need to examine how the cloud diurnal cycle changes below or within aerosol layers and their radiative effects. Understanding how these relationships vary as the biomass burning season progresses are also critical in the context of aerosol-cloud climatology. Although SEVIRI is a multispectral sensor that possesses a characteristically high repeat cycle, it lacks adequate solar channels to retrieve optical and microphysical properties of AACs at a MODIS capability.

Finally, model-observation inter-comparison projects will consolidate the current progress of aerosols and improve aerosol forecasting at various time scales. Observations of all sources should be addressed including in situ, ground, and satellite observations. Figure 5.1 shows a preliminary AOD comparison between 4STAR and WRF-CAM5 during ORACLES 2016. These projects will improve our understanding of all-sky aerosol direct radiative effects.
Figure 5.1. AOD comparison between 60-second averaged AOD from 4STAR and WRF-CAM5 AOD during August and September 2016 (Shinozuka et al., 2018).
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