CALIPSO-Derived Three-Dimensional Structure of Aerosol over the Atlantic Basin and Adjacent Continents

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ABSTRACT

Accurate modeling of the impact of aerosols on climate requires a detailed understanding of the vertical distribution of aerosols. The Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) provides continuous high-resolution vertical profiles of aerosol properties on a near-global scale. Here the CALIPSO Vertical Feature Mask is used to document the three-dimensional (3D) frequency-of-occurrence distribution of aerosols over a broad region of the Atlantic Ocean, Africa, Europe, and the Americas. The 3D distributions illustrate the seasonal cycle in the zonal and meridional variability of the vertical profiles of mineral dust, biomass-burning smoke, polluted dust (external mixture of dust and smoke), and polluted continental aerosol, and also of their emissions sources and transport pathways. Four aerosol domains stand out in the product: dust over North Africa and the Middle East and smoke over southern Africa and South America. The transport pathways of African dust and smoke over the Atlantic are evident. The intertropical convergence zone (ITCZ) plays a clear role in limiting the southward transport of North African dust and northward transport of South African smoke. Dust and smoke are mixed in the ITCZ and consequently the highest probability of polluted dust is found there, even though the probabilities of dust and smoke in this region are relatively low. The mixing of dust and pollution has significant implications for cloud microphysical processes over a broad region of the Atlantic.

1. Introduction

Natural and anthropogenic aerosols play important roles in Earth’s climate by altering the radiation budget and influencing key processes in the hydrological cycle. Aerosols affect climate directly by scattering and absorbing solar and longwave radiation (Forster et al. 2007). Aerosols indirectly affect climate in two ways. The first indirect effect refers to the increase in cloud condensation nuclei (CCN) due to increasing aerosol, which results in decreased cloud droplet size, increased number concentrations, and thus increased cloud albedo (Twomey 1977). The second refers to the role of aerosols, acting as CCN and ice nuclei (IN), in affecting changes in cloud lifetime due to decreased precipitation efficiency (Albrecht 1989). Based on the 2007 Intergovernmental Panel on Climate Change (IPCC) report, estimates of the direct radiative effect vary between +0.61 and +2.43 W m$^{-2}$ and between −0.22 and −1.85 W m$^{-2}$ for the cloud albedo effect. The impact of aerosols on the hydrological cycle suffers from even greater uncertainties (Forster et al. 2007). Reducing the uncertainties in aerosol climatic effects is a necessary step toward increasing the reliability of global climate models in reproducing the current climate and projecting into the future.

Global and regional circulation models are typically used in concert with in situ and remote sensing measurements to improve estimates of aerosol effects (Penner et al. 1994; Kaufman et al. 2002; Ming et al. 2005; Ming and Ramaswamy 2009). Numerous direct radiative effect studies using model simulations typically calculate radiative fluxes at the top of the atmosphere and at the surface using various model estimations for preindustrial and postindustrial aerosol loading (Chung and Seinfeld 2005; Myhre et al. 2007; Shell and Somerville 2007). Results are often inconsistent in the magnitude and, in some cases, the sign of the radiation fluxes (Forster et al. 2007; Schulz et al. 2006; Yu et al. 2006). Several sources of error contribute to these discrepancies, the most dominant being the lack of accurately resolved aerosol vertical distributions (Claquin et al. 2007).
of the four types. section 2 contains the descriptions of the data used in this study and data processing methodology. Results are introduced in section 3, followed by conclusions in section 4.

2. Data and methods

a. CALIPSO satellite data

The National Aeronautics and Space Administration (NASA) and the Centre National d’Etudes Spatiales (CNES) launched the CALIPSO satellite into the A-Train satellite constellation on 28 April 2006. The payload of CALIPSO includes a high-resolution Cloud–Aerosol Lidar with Orthogonal Polarization (CALIOP). It consists of a dual-wavelength polarization lidar (532 and 1064 nm) in concert with three receiver channels that measure the backscatter intensity at 532 nm, polarized 32 nm, and 1064 nm wavelengths with a pulse energy of 110 mJ/channel and repetition rate of 20.25 Hz (Hunt et al. 2009). One of its products is the VFM. The VFM algorithm is described in the document CALIOP Algorithm Theoretical Basis Document, Part 3: Scene Classification Algorithms (Liu et al. 2005). It identifies the presence of clear air, cloud type, or aerosol type at each horizontal and vertical grid point. Areas of enhanced scattering are first evaluated by the Scene Classification Algorithm (SCA), which evaluates if the area is nonclear-air feature, the surface, totally attenuated, and if the feature is elevated. Upon detection of a feature, the SCA identifies the layer as cloud or aerosol based upon a confidence function, computed from the layer mean value of the attenuated backscatter coefficient at 532 nm, the ratio of the backscatter at 1064 nm and 532 nm (volume color ratio), the height of the center of the layer, and an existing probability density function (PDF) database. Once the SCA determines that the feature is aerosol, the aerosol is ascribed to one of seven aerosol types based upon extinction-to-backscatter ratio estimated from input parameters including backscatter coefficient measurements, depolarization ratio, altitude, location, and surface type.

The seven aerosol types defined in this analysis scheme are as follows: desert dust (henceforth Dust), biomass burning smoke (soot and organic carbon; Smoke), polluted dust [externally mixed Dust and Smoke (PDA)], clean continental aerosol (background aerosol; sulfates, nitrates, organic carbon, and ammonium), polluted continental aerosol [background aerosol with urban pollution (PCA)], marine aerosol (sea salt), and “other”; the last is currently not in use (Liu et al. 2005). Our study focuses on four aerosol types: Dust, Smoke, PDA, and PCA. Capitalization of aerosol
type hereafter indicates CALIOP derived aerosol type. We exclude the other three aerosol types (clean continental, marine, and “other”) because the temporal–spatial patterns were noisy and did yield coherent patterns. We use Level 2 VFM Validated Stage 1 Version 3 data in this study.

The accuracy of CALIPSO VFM aerosol identification is generally high, particularly for Dust and PDA classification (Mielonen et al. 2009) but is prone to misclassification errors under certain circumstances. The most prominent error is the misclassification of Dust or Smoke as cloud, which occurs when the dust or smoke layer is thick, and optical properties become similar to those of cloud, or if the aerosol is located near a cloud layer (Z. Liu et al. 2009). Z. Liu et al. (2009) found that this type of error occurs in less than 1% of the cases in their study involving Dust; they suggest that the Smoke misclassification error is much less. However the misclassification of Smoke as PDA can also be an issue (Mielonen et al. 2009), particularly in the boundary layer (Omar et al. 2009). Misclassification of aerosol under thin ice clouds can also be a problem (Z. Liu et al. 2009). These errors should be borne in mind when results from this study are interpreted.

The analysis period of this study includes all available VFM data in the period 16 June 2006–16 June 2010, which encompasses four complete seasonal cycles. Boreal summer [June–August (JJA)] includes June, July, and August 2006–09 with the addition of 1–16 June in 2010 to complete the season because the 2006 dataset began on 16 June. Fall [September–November (SON)] includes September, October, and November 2006–09. Winter [December–February (DJF)] includes December, January, and February 2006/07–2009/10. Spring [March–May (MAM)] includes March, April, and May 2007–10. We use only nighttime data (descending orbits) because the returns are of significantly better quality than during daytime when the signal-to-noise ratio is reduced by an effect of the solar background (Powell et al. 2009). Horizontal and vertical spatial resolution varies with height. In this study, only the lower 8.17 km of the troposphere is examined. For these altitudes, the VFM has a vertical resolution of 30 m and horizontal resolution of 333 m.

b. Meteorological data

Tropical Rainfall Measuring Mission (TRMM; Kummerow et al. 1998) 3B42 precipitation data were used to describe the location of the ITCZ. This is a TRMM-adjusted merged-IR precipitation estimate (Kummerow et al. 2000). The gridded daily data (0.25° × 25° resolution, 50°N–50°S) cover the analysis period described above. Some plots include wind from European Centre for Medium-Range Weather Forecasts (ECMWF) Interim Re-Analysis data (ERA-Interim; Simmons et al. 2006). The gridded (2.5° × 2.5° resolution) daily data cover the described analysis period.

c. Seasonal averaged occurrence probability

VFM Level-2 swath data were gridded into a three-dimensional latitude, longitude, and height matrix with dimensions of 1° × 1° × 30 m for 290 levels with a range of −0.5 to 8.17 km. The rectangular domain of interest covers 40°S–60°N and 60°E–100°W. This domain includes regions that are known for their major aerosol sources or are influenced by high concentrations of transported aerosols: Africa, the Atlantic Ocean, South America, Caribbean, and the eastern United States. There are 161 × 101 × 290 horizontal and vertical grid points in this 3D domain. At each grid point, we tallied the occurrence of Dust, PDA, PCA, and Smoke using the entire swath (a meridional cross section at 20°E is shown in Fig. 1a). The total occurrence number of each type of aerosol was then normalized by the total number of valid satellite passes at each grid point (Fig. 1b; same cross section as 1a). A valid satellite pass is defined here as a VFM indication of clear air, aerosol, or cloud.

We identified the number of valid satellite passes in the grid box containing the greatest number of passes in the domain. We only retained the averaged data for grid points where the number of valid satellite passes exceeded 15% of that number. This criterion is established to minimize the spread of anomalous aerosol identifications via running mean averaging. Figure 1c illustrates such anomalous identifications. A grid point with a low number of satellite passes can nonetheless yield a high FoO if most of the passes identify aerosol. It is clear from the figure that this is not accurate. Including only the points with a larger number of passes (exceeding the 15% threshold) helps to remove the anomalous vertical lines of constant FoO. The number of valid satellite passes is highly variable horizontally and vertically, with greatest values at upper altitudes as optically thick cloud cover and precipitation limit the number of observations below (maximum approximately 7500 in a season at a grid point, and minimum approaching zero near the surface).

The normalized and filtered data were then smoothed to highlight coherent large-scale structures (Fig. 1d). First, we applied a 13-point latitudinal running mean; for the grid points near the boundaries, we incorporated data outside the analysis domain. A second smoothing was applied using a 3-point longitudinal running mean in a similar manner. Results remain the same if the running mean is applied to longitude and 3-point running mean to latitude.
The thus-processed data yield a 3D structure of FoO. Here we visualize the data in meridional-vertical cross sections at every 10° longitude. An example is Fig. 2, which shows the summer mean structure of FoO for total aerosol. As previously stated, probabilities less than 15% are not plotted so that a cohesive structure can be more clearly observed. Hereafter, such a structure of occurrence probability will be referred to simply as a FoO distribution or structure.

d. Seasonal cycles of occurrence probability

In addition to seasonal averages, we produced a 3D seasonal cycle of FoO that yields a zonally integrated 3D structure as a function of latitude, height, and time. We tallied the VFM data into a daily 3D array at each latitude (1°), longitude (1°), and height (30 m) over the same analysis domain. Occurrences in longitude \(x\) are summed and normalized by the total valid satellite passes at the same grid point, and variable \(N\) is the number of grid points in a specified longitudinal range. The 3D dataset is then smoothed twice using a 29-day running mean and once using a latitudinal three point running mean. There are only small numbers of valid satellite passes at each grid point for the seasonal-cycle structures (365 \(\times\) 101 \(\times\) 290 versus 166 \(\times\) 101 \(\times\) 290), and we found it unnecessary to impose a threshold for satellite minimum passes as we did for the seasonal averaged structure. As in the seasonal average plots, probabilities less than 15% are not shown.

The ITCZ location was estimated using TRMM estimated precipitation. TRMM daily data were regridded into a 1° \(\times\) 1° \(\times\) 365-day matrix to match the resolution of FoO structure and then averaged over the same longitudinal range (see section 3e). A least squares spline fitting was applied to zonally averaged TRMM rainfall maxima as a proxy of the latitudinal position of the ITCZ.

3. 3D aerosol structures

We find significant seasonal variations in 3D aerosol structures in such features as source regions; transport patterns; vertical, longitudinal, and latitudinal distributions; and the locations and sizes of most active aerosol...
regions. In this section, we summarize some of the major features of these 3D aerosol structures.

The Total Aerosol 3D FoO distributions for the months of JJA are shown in Fig. 2 (top panel). In the bottom panel, each figure shows vertical profiles along longitudes at 10° intervals. The vertically averaged FoO values are projected on the horizontal plane. The averaged vertical profiles across the latitudes are projected on the panel at the right of the domain and the averaged profiles along the longitudes are shown on the back plane. This plotting convention is used in other figures.

In this work we see evidence of strong aerosol activity throughout much of the domain both in the Northern and Southern Hemispheres. As we will show in later sections, JJA are the months of maximum activity, followed in order by SON and MAM. Total Aerosol shows the least activity in DJF. Our 3D structures display features that have implications about source–transport relationships. For example in Fig. 2, we can identify four distinct regions with high aerosol probabilities: West Africa and the Atlantic corridor of the Saharan air layer (SAL; Carlson and Prospero 1972), southern Africa, South America, and the central and eastern United States. Over Africa and the Americas we see a clear demarcation in the north-south profiles in the vertically averaged FoO. This demarcation can be seen in the pattern projected on the base of the top panel and also in the 10° longitudinal-vertical cross sections shown in the bottom of the figure. We will show that these demarcations are due to the effects of the ITCZ as seen in the zonal mean (pattern projected on the right-sidewall of the top panel) as well as along each longitude. Near the western boundary of the domain, in the Northern Hemisphere, there are two distinct aerosol regions—one over the Caribbean and another over the central and eastern continental United States. We will show that this reflects two types of aerosol: dust over the Caribbean originating from Africa (Prospero and Lamb 2003) and smoke presumably originating from fires and possibly combustion pollutants over the United States. Also, the Caribbean aerosol FoO structure is clearly distinct from the region of high FoO values over South America. There is a clear east–west separation between the Southern Hemisphere sources in Africa and those in South America; this demarcation is mainly due to the distinct character of these two source regions and a lack of efficient zonal transport mechanisms between them.
In the Northern Hemisphere, the regional associations can be best seen in the meridionally averaged FoO pattern (projected on the back wall of the top panel). We will show that the four sections of high FoO are due predominantly (from right to left) to Middle Eastern dust, African dust, South American smoke, and eastern U.S. smoke.

Other details are apparent in the 3D structure. Maximum aerosol height is different over northern and southern Africa and between Africa and Americas. There is a sharp cutoff in aerosol probability at all altitude levels over West Africa at the southern boundary and over southern Africa at the northern boundary. In addition, we see on the back panel an "aerosol anvil" between about 20°W and 10°E. We discuss these and other features in detail in the rest of this section.

a. Dust

In JJA (Fig. 3a), several distinct dust source regions can be seen. The primary dust source activity is located in North Africa; emissions from these sources extend westward over the tropical Atlantic Ocean to the Caribbean. Another region of high dust occurrence is located over the Middle East. This source is distinct from the North African sources as is evident in the meridional average projected on the back plane. In SON (Fig. 3b), this separation remains identifiable although less distinct. It is no longer present in DJF (Fig. 3c) when dust activity in general is at a minimum, but it reappears in MAM (Fig. 3d) albeit with a less distinct boundary than summer. The two isolated regions of high FoO are linked to two distinct dust sources. The more western feature is linked to the well-known Saharan–Sahelian source complex (Middleton and Goudie 2001; Prospero et al. 2002; Engelstaedter et al. 2006); the eastern feature is linked to sources in the Arabian Peninsula and the Middle East (Prospero et al. 2002; Washington et al. 2003). The apparent separation of source region is due to the seasonal variability of dust FoO and to the prevailing wind climatology. Northerly wind with relatively

Fig. 3. Dust FoO in JJA, SON, DJF, and MAM, with the same plotting convention as in Fig. 2.
pristine air at 20°–40°E and 25°–35°N dominates in JJA, essentially slicing into the FoO structure. In DJF, the northerly winds are still present at the lowest levels, but the high FoO of dust is shifted south of its summer location so that the separation is not apparent in the meridional average.

Other dust sources are seen in Fig. 3 but they are much weaker. Weak but persistent activity is visible in southern South America, particularly in Argentine Patagonia (Prospero et al. 2002). This region is believed to be the major source of dust that is transported to the South Atlantic (Gasso and Stein 2007). It is notable that dust is confined to low altitudes. Two very small and weak sources are located in southern Africa in Namibia and Botswana, consistent with previous studies (Prospero et al. 2002; Bryant et al. 2007). Both are visible, albeit very weakly and at low altitudes, throughout the year. There also appears to be minor dust activity over the central United States, especially in SON and MAM, which is consistent with dust activity in this region (Orgill and Sehmel 1976). The vertical extent of Dust over all these weak source regions is limited to low altitudes (< 2 km).

The westward transport of dust from North Africa undergoes substantial seasonal changes in FoO structure and geo-spatial extent. During JJA, Dust extends across the Atlantic Ocean and into the Caribbean as far as 100°W (consistent with Prospero and Lamb 2003; Vuolo et al. 2009) and as far north as south Florida, 30°N (as in Prospero 1999). Over the Sahara and, indeed, over most of North Africa, the FoO structure shows a rather uniform maximum altitude of 6 km; there is a well-defined maximum FoO core that extends from near ground level to about 5 km with FoO values near 75% or above. Over the Atlantic FoO varies from above 65% in the core at 20°W to less than 30% at 80°W; however, the center of the core remains essentially unchanged at approximately 3.5 km. The vertical extent gradual decreases from about 6 km near the West African coast (in line with Léon et al. 2009) to less than 4 km over Central America (like Ben-Ami et al. 2009; Vuolo et al. 2009). The meridional coverage of Dust also decreases from the east to west.

In SON (Fig. 3b), the maximum altitude over much of North Africa is relatively constant at about 5.5 km, but the core with FoO >75% extends only to 2 km. The vertical extent of dust is less than in summer almost everywhere, as is evident in the meridional average. The westward extent of dust FoO is greatly reduced; a coherent structure is visible only to about 50°–60°W and the core FoO is only about 15%. Similar to the previously noted feature in JJA, the SON FoO meridional averages projected on the back plane show a clear demarcation of the same previously-seen two regions of maximum dust probability, one over North Africa and the other over the Arabian Peninsula and the Middle East.

In DJF (Fig. 3c), dust activity is seen to be at an annual minimum throughout the domain and the vertical distribution and spatial coverage are also greatly reduced. Across much of North Africa, the dust maximum altitude is near 4 km; along the coast of Africa, measurements show the top between 3 and 4 km (Léon et al. 2009; Vuolo et al. 2009). While dust activity continues over North Africa, it is concentrated in the lower latitudes in the Sahel–Soudano region where the Bodélé Depression in Chad, one of the world’s most intense dust sources, is known to be most active in this season (Washington et al. 2003). Dust activity over the Arabian Peninsula and the Middle East has largely ceased. Over North Africa, the maximum altitude decreases to about 3 km with a core FoO of 75% at 1.5 km. Westward dust transport over the Atlantic is at a minimum, with an abrupt reduction in the FoO from 50% at 20°W to 15% at 30°W. The top of the Dust FoO is remarkably zonally uniform across all of North Africa almost to the westernmost extent of transport in Fig. 3c. Meridionally, FoO exhibits a bimodal pattern with lower maximum altitudes north of 20°N and greatest and highest-altitude FoO in the Sahel–Soudano region, (consistent with Ben-Ami et al. 2009). In contrast to JJA and SON, the greatest FoO values are found in a layer that is rather shallow, about 1 km. Over the Atlantic, the meridional coverage shifts southward relative to JJA and SON, presumably linked to the seasonal shift in the large-scale circulation and to the southward movement of the ITCZ.

In MAM (Fig. 3d), dust activity increases sharpenly over North Africa, the Arabian Peninsula, and the Middle East with FoO values and altitude levels approaching those in the summer season. Indeed, the Dust FoO across this region is remarkably uniform with only a subtle indication of a demarcation in sources from the west coast of Africa to the eastern boundary at 60°E. However, in contrast to JJA, the axis of trans-Atlantic transport is located further to the south, extending to the northeast coast of South America (in agreement with Prospero et al. 1981) and reaching into the Amazon Basin (consistent with Swap et al. 1992). Some dust activity is also seen in the central United States.

CALIOP clearly shows that dust is present in and across the ITCZ vicinity in MAM, most notably over the eastern tropical Atlantic (Adams and Zhang 2012, manuscript submitted to Bull. Amer. Meteor. Soc.); note that some cross-ITCZ FoO was also observed in DJF (Fig. 3c). This is possible because there is only
intermittent ITCZ rainfall and because of the presence of strong, shallow northerly flows (immediately above the boundary layer) acting to transport dust southward over cloud in DJF and MAM.

From a comparison of Fig. 3a, which shows summer Dust FoO, and Fig. 2, which shows total aerosol for that season, it is clear that north of the equator Dust is by far the dominant contributor to Total Aerosol. In contrast, Dust makes a negligible contribution to Total Aerosol south of the equator where Smoke dominates, as will be discussed next.

b. Smoke

The 3D structure of Smoke FoO reveals four distinct source regions in the analysis domain: southern Africa, South America, the southeastern United States, and Europe, all of which vary seasonally (Fig. 4). Of these, the most robust and seasonally persistent sources are located in southern Africa, where biomass burning is known to be intense and extensive (Cooke et al. 1996; Hao and Liu 1994). Smoke activity is greatest (FoO 75%–80%) in JJA and SON and is weakest (FoO <40%) in DJF, consistent with burning estimates (Ito et al. 2007; Chang and Song 2009). However, CALIOP seems to underestimate the frequency of burning in the Sahel–Soudano region where fire is known to be widespread in DJF (Kaiser et al. 2011). The underestimate is most likely due to the mixing of smoke with dust, which is widespread in this region at this time of year (Fig. 3c) so the expected Smoke FoO appears only as PDA (see below). It is notable that, if biomass burning is the main source of smoke over southern Africa, our data suggests that burning is active all year round, including the local rainy season (DJF). Smoke reaches the highest altitudes (5–6 km) in SON and lowest in DJF and MAM (<5 km). It propagates westward into the Atlantic Ocean (~30°W) only in JJA and SON, consistent with the distribution and seasonality of CO over this region (Edwards et al. 2006). As seen on the backplane in Figs. 4a and 4b, transport takes place in an elevated layer between 3–6 km, consistent with measurements made in past field campaigns.

![Fig. 4. As in Fig. 3 but for Smoke.](http://journals.ametsoc.org/jcli/article-pdf/25/19/6862/3999030/jcli-d-11-00672_1.pdf)
c. Polluted continental aerosol

PCA is widespread throughout the domain but it is primarily confined to low altitudes (Fig. 5). As we might expect, pollution is present in all seasons over North America and Europe. However, the highest FoO values and the greatest variability are seen over South America and southern Africa. In some areas (e.g., in SON over Brazil) it is clearly associated with smoke and may be a miscategorization. But in other areas, in particular southern Africa but also to a lesser degree in South America, the structures are distinctly different than that of Smoke FoO. For example, the largest PCA FoO values are at the boundaries of the Smoke structure over southern Africa, not collocated. In the CALIOP Scene Classification Algorithm (SCA), the only difference between a resulting Smoke and PCA classification is that Smoke is an elevated layer and PCA is a non-elevated layer, regardless of the thickness of the layer; the other aerosol type defining characteristics (e.g., depolarization ratio and integrated backscatter) are identical. Therefore, near surface smoke layers are classified as PCA over land and ocean.

North of the equator, there is little evidence of transport of PCA from the continents to the oceans except perhaps for some transport off the east coast of the United States. Over southern Africa, the pattern of FoO is essentially unchanged across the seasons. The greatest values are located near and on the coasts in regions where the population density is relatively high and where one might expect anthropogenic emissions to be high. In contrast, Smoke activity is centered well inland (Fig. 4). The difference between the Smoke distributions and that of PCA is especially notable in DJF (and to a lesser extent in MAM) when smoke activity is weak. The FoO distributions suggest that there is strong transport to the Indian Ocean in all seasons. Nonetheless, it is debatable whether PCA FoO in this region is indeed anthropogenic emissions or biomass burning smoke because of the classification method.

Similarly, in South America the regions with highest FoO of PCA are found along the coasts extending from the Caribbean to northeast South America and south to Argentina. In JJA (Fig. 5a) and SON (Fig. 5b), the PCA regions merge with those of Smoke. Indeed the location of the largest PCA FoO values and the highest vertical distributions are seen in central Brazil in the same region where the greatest smoke activity is found (Fig. 4). The greatest FoO is in JJA (Fig. 5a) and SON. Throughout the year, PCA is transported from Brazil and Argentina eastward over the Atlantic; this transport is not seen in the Smoke structures.

In Europe, PCA is present in all seasons and covers almost the entire region from the Atlantic to the eastern bound of the domain. In JJA and MAM, PCA and Smoke are collocated over central Europe although PCA coves a much larger area than does Smoke. The greatest PCA FoO in Europe occurs in SON (Fig. 5b) and DJF (Fig. 5c).

The eastward propagation of U.S. pollution is evident in JJA, SON, and MAM, but it is spatially limited, FoO values are very low, and, by definition, confined near the
surface. Although PCA is defined in CALIOP documentation as urban pollution and Smoke is defined as a biomass burning smoke (Liu et al. 2005), a comparison of Fig. 4 and Fig. 5 suggests that it may be beneficial to consider PCA when evaluating smoke using CALIOP products.

d. Polluted dust aerosol

The distribution of polluted dust (PDA, Fig. 6) is markedly different from the other species considered here. Most notably, detectable amounts of PDA are seen over a much larger area than the other species, essentially covering the entire continental areas and extending over large areas of ocean. Over most of this huge area, PDA values are low and generally confined to low altitudes with some significant exceptions. In addition, the areal coverage does not change much with season.

In JJA (Fig. 6a), there are two regions where PDA FoO values are large and reach substantial altitudes. One region extends from the Mediterranean basin into Russia, likely due to northward advecting dust mixing with European pollution and smoke. The second is centered over the Caribbean and extends eastward. The Caribbean PDA is clearly linked to the mixing of African dust with smoke and pollution most likely emitted from regional sources. On Barbados (13.18°N, 59.43°W), which underlies the core of the dust plume, about half of the nonsea-salt sulfate mass is anthropogenic (Savoie et al. 2002) and 85% is in the fine particle mode (Li-Jones and Prospero 1998) that, when mixed with dust, is consistent with the CALIOP identification as PDA. In contrast, over West Africa and eastern Atlantic PDA FoO values are very low in JJA. Over the western region there is a clear difference between the FoO of Dust and that of PDA in that the Dust distribution is seen as an elevated layer (consistent with the presence of the SAL) while PDA is mostly concentrated below the SAL altitudes with the highest values in the marine boundary layer. The Caribbean PDA distribution merges with, but is distinct from, increased PDA FoO seen in the central
and eastern United States and, to the south, over central Brazil.

The high PDA FoO values observed over the Mediterranean basin in JJA are consistent with the transport and mixing of African dust and European pollutants (Levin et al. 1996; Lelieveld et al. 2002). The most intensive and extensive PDA FoO values and highest mixing depths are seen through the Middle East, across eastern Europe and into western Russia where there are many dust sources around the Black, Caspian, and Aral Seas (Prospero et al. 2002).

In SON (Fig. 6b), the overall distribution of PDA FoO does not change much but the areas of greatest activity shift. The PDA from mixing of African dust and pollution is no longer visible but increased FoO and mixing depth are seen across northern South America, clearly the result of the mixing of dust with regional smoke/pollution along the northern boundary of the ITCZ. Over the eastern domain, increased PDA is seen over the Middle East and especially the Saudi Arabian Peninsula where PDA is mixed to 4–5 km. Here, too, it is notable that the greatest mixing depths occur along the northern edge of the ITCZ.

The DJF FoO (Fig. 6c) shows a wide corridor (≈20° of latitude) of high values that extends from East Africa to the Caribbean and which precisely straddles the ITCZ over much of its length, reaching altitudes of ≈4 km. As stated in earlier sections, DJF is the burning season in the Sahel-Soudano region and there is also intense dust activity. The mixing of dust and smoke (and possibly pollution) over the African Sahel–Soudano region is well documented by research flights (Kim et al. 2009; Matsuki et al. 2010). As discussed in a later section, dust generated over the Sahara is apparently advected into the ITCZ region where it mixes with smoke, subsequently detected by CALIOP as PDA. This would explain why CALIOP does not see smoke during the DJF (Fig. 3c) as previously discussed. The presence of high concentrations of these two aerosol species—one as fine particles and one as coarse—in the ITCZ over such
a large area may have a significant impact on cloud microphysics across this region (Huang et al. 2009a).

In MAM, dust transport across the Atlantic is evident (Fig. 3d). In Fig. 6d we once again see the colocated dust with pollution, resulting in FoO distributions similar to JJA but generally weaker. Polluted dust no longer straddles the ITCZ with the exception of the slice at 30°W. Large values of PDA FoO are present over the Arabian Sea, evidently from the mixing of pollutants borne with Arabian and African dust during the winter monsoon.

e. Dust and smoke comparison with polluted dust aerosol

Dust, Smoke, and the combination of the two (PDA) are clearly the dominant aerosol species over the study domain during JJA. In Fig. 7, we show the combined FoO for these species on the latitude–height slices in JJA. These are useful in characterizing the longitudinal and latitudinal range of each individual aerosol type and they can provide insights to the possible controlling mechanisms of each type of distribution and the conversion from pure Dust or Smoke to PDA. Because Smoke and Dust structures have been shown previously and trans-Atlantic transport of smoke is limited to JJA and SON, only the summer season of combined Dust and Smoke is included.

It is evident from Fig. 7 that there is overlap between Dust and Smoke. Over land (Fig. 7a), three regions of Dust/Smoke overlap are evident: the Mediterranean and southern Europe, the ITCZ region (where ascending motions are), and southern Africa. European smoke and pollution advects southward where it mixes with dust (Lelieveld et al. 2002). African dust is also on

FIG. 7. JJA vertical cross sections of aerosol FoO with wind vectors averaged over (a) 11°–40°E, (b) 16°W–10°E, (c) 34°–17°W, and (d) 80°–35°W. Vertical wind amplitude is amplified by a factor of five for clarity. Gray shading represents PDA, blue contours Dust, and green contours combined Smoke and PCA. Contours are plotted in 10% intervals with the exception of the first contour at 5%. Here, 30% contour is dashed for reference.
occasion advected deep into central Europe (Stuut et al. 2009). In Fig. 7a, CALIOP shows the presence of Dust FoO greater than 5% as far north as ~50°N. The mixing of pollution and dust results in an area of large PDA FoO that extends southward to ~20°N. At the Dust/Smoke boundary, between 0° and 10°N, the apparent presence of PDA is probably the result of the mixing of dust and smoke across the ITCZ (Mari et al. 2008; Matsuki et al. 2010). Analysis of the mixing mechanisms associated with this “leaky barrier” is described in Adams and Zhang (2012, manuscript submitted to Bull. Amer. Meteor. Soc.). Over southern Africa, wind vectors indicate the downward advection of smoke into the weak southern African dust sources. It is unclear how dust in this area is elevated up to 3 km to create PDA, especially when we would expect to see very little dust. It is possible that the PDA FoO from the ground to 3 km is due to misclassification of Smoke as PDA, but it is more likely that the upward component of the wind is lost in the zonal averaging (Adams and Zhang 2012, manuscript submitted to Bull. Amer. Meteor. Soc.).

Farther west (Fig. 7b), where the northern section is still over the aerosol sources but the southern portion is over ocean, we see a continued overlap between European smoke and Saharan dust, but only within the outer contour of each distribution (FoO between 5%–10%); PDA is more frequent than Dust or Smoke in the area. The elevated Smoke and Dust structures continue to mix in the ITCZ region, but the mechanism to create PDA is different; the mixing is due to the shallow meridional circulation (Zhang et al. 2006, 2008; Adams and Zhang 2012, manuscript submitted to Bull. Amer. Meteor. Soc.), which transports PDA downward. Although the source is weak, southern African dust is transported westward only as PDA.

Over the open Atlantic (Fig. 7c), the PDA FoO structure is similar to the previous one. European smoke is no longer present; a small FoO of PDA still exists between 30° and 40°N. An additional meridional circulation north of the ITCZ is present; this acts to collocate the Dust and PDA into a cylindrical area for the remainder of the westward transport.

South American Smoke and Dust aerosol converge only over the southern dust source (Fig. 7d). Descending motion in this area acts to transport smoke downward where it mixes with dust, creating PDA. Near the equator, there is little interaction between Dust/PDA transported over the Atlantic and Smoke of South American origin. It is likely that most of the PDA north of 5°N contains smoke with African origin. The other three seasons (not shown) exhibit similar Dust/Smoke conversion patterns, though not as clearly defined as in JJA. The lack of a coherent Smoke propagation signal over the Atlantic in DJF and MAM can be attributed to mixing over a smaller horizontal distance; the Smoke and Dust mix to PDA east of 10°E rather than 20°W as in JJA and SON.

f. Seasonal migration

In Fig. 8 (top), we show the continuous seasonal cycle of the FoO of combined Dust and Smoke zonally integrated over the eastern side (60°E–31°W) of the previously used analysis domain. The FoO of the region south of the ITCZ is predominantly Smoke; the FoO north of the ITCZ is predominantly Dust. Maximum heights of Dust and Smoke in the seasonal cycle are consistent with maximum heights seen and described previous sections.

Dust and Smoke are observed in areas of minimum rainfall, overlapping only in late summer and early fall. The time-space area of observed Smoke south of the ITCZ occurs in an area of rainfall minima. The northerly extent of Smoke increases from 10°S in June to 5°N in mid-September then retreats southward in early November. This closely tracks the annual migration of the ITCZ. The FoOs of Dust are greatest and most persistent over the year in an area of rainfall minima north of the ITCZ. Along 20°N, dust activity persists all year long with a maximum in the summer months. The southern boundary of the Dust FoO tracks the movement of the area of rainfall associated with the ITCZ.

The temporal–spatial distribution of PDA FoO (Fig. 8, bottom) is quite different from those of Dust and Smoke. The largest FoOs of PDA are located in the vicinity of the ITCZ, but with an inverse relationship to rainfall amount: in DJF when rainfall rates are decreased, PDA FoOs are at a maximum; when rainfall is more abundant in JJA, PDA FoOs are at a minimum. Despite the inverse relationship, the seasonal migration of PDA closely resembles maximum rainfall seasonal cycle. There is also a marked difference in the altitude distribution of PDA compared to Dust and Smoke; this altitude changes greatly over the course of the year. These relationships are best seen in the seasonal profiles along a specific latitude as depicted in the bottom small panels of Fig. 8. For example along 20°N, Dust and Smoke are present all year long at maximum altitudes ranging from 2 km (DJF) to 6 km (JJA). In contrast, PDA is confined to low altitudes, a maximum of 1.5–2 km in DJF and a vanishingly small minimum in JJA. The great temporal and spatial variability across this region is illustrated by comparing the 20°N distribution with those at 10°N where the intrusion of the monsoon circulation greatly reduces dust activity at midyear and where, conversely, PDA is present much of the year and persistently attains a maximum altitude of about 4 km.
4. Summary and conclusions

Using the CALIPSO Vertical Feature Mask, we created 3D representations of climatological aerosol occurrence probabilities over the Atlantic basin and the surrounding continents to gain a better understanding of the temporal and spatial variability of different types of aerosol. Our analysis includes the relationship of aerosol distributions to the location of the ITCZ based on TRMM rainfall maxima estimates. Our results are consistent in many cases with previous studies with more local focuses. This is the first time the 3D structure and variability of aerosol are viewed over the entire region of the tropical and subtropical Atlantic Ocean and its adjacent landmasses.

Within our domain, the distribution of Total Aerosol is chiefly determined by the presence of either Dust or Smoke or the combination of the two. Polluted Continental Aerosol, although widespread, generally is present at low frequency of occurrence values and, by VFM definition, it is confined to low altitudes. In contrast during active dust and burning seasons, Smoke and Dust
are often mixed and transported to altitudes of 4–6 km. Well-known source regions of dust and smoke are clearly apparent in the 3D seasonal occurrence structures. The Sahara Desert and the Middle East stand out as the most intense and most persistently active dust source regions. There are a few scattered minor sources in southern Africa and southern South America; the frequency of occurrence values over these sources are very low and the Dust is generally confined to shallow near-surface layers. Our Dust source distribution and activity results are consistent with previous studies based on various satellite products (e.g., Prospero et al. 2002; Formenti et al. 2011) and other metrics of dust activity (Goudie and Middleton 2006).

The dominant Smoke source regions are located in South America and in equatorial and southern Africa, consistent with previous studies of biomass burning activities. Evidence of weaker sources is seen in the eastern United States and Europe. The CALIPSO Smoke product does not show smoke in the Sahel in DJF, known to be the burning season. Instead, smoke shows up as Polluted Dust, a consequence of the strong mixing of dust and smoke in this complex monsoonal mixing regime.

The probability of the westward transport of Dust and Smoke and their vertical extent vary with season; the furthest westward transport and highest frequency of occurrence values occur in JJA and MAM. In general, based on the CALIPSO product, smoke does not appear to be transported great distances compared to dust. This difference may be an artifact of the VFM, it may reflect the differences in meteorological conditions associated with the smoke and dust events or in the removal process (e.g., possibly linked to the very different properties of smoke and dust with respect to cloud nucleating processes), or it may be influenced by the injection height. Smoke and Dust frequency of occurrence spatial and temporal structures are consistent with observed values in other studies (e.g., Prospero and Lamb 2003; Edwards et al. 2006; Leon et al. 2009; Vuolo et al. 2009).

The ITCZ clearly plays a major role in affecting dust and smoke distributions throughout the year, limiting the southern extent of Dust from North African sources and the northern extent of Smoke from southern Africa. The larger-scale wind fields associated with the ITCZ appear to play a role in the mixing of dust and smoke. In the ITCZ vicinity, Dust and Smoke frequency of occurrence are at a minimum, while the probability of Polluted Dust is at a maximum. The most interesting case is in DJF when the Polluted Dust structure straddles the ITCZ over the entire span of the Atlantic. This is due to the initial rapid mixing of dust and smoke near the source regions at which time Polluted Dust reaches its maximum frequency of occurrence and highest altitudes.

There are some limitations of the CALIPSO product related to aerosol identification that could affect our interpretations. Polluted Dust is defined as a mixture of Dust and Smoke based primarily on the depolarization ratio. It is not possible to ascertain the relative contribution of each aerosol that would be required to yield this classification. It is notable (Fig. 6) and somewhat surprising that Polluted Dust is by far the most widespread aerosol type, blanketing essentially all the continental areas and a large area of the Atlantic in all seasons. However, over most of the Atlantic (e.g., the mid-to-high latitudes of the North Atlantic and most of the South Atlantic), dust concentrations are known to be very low, typically a few tenths of $\mu g \ m^{-3}$ (Arimoto et al. 1995; Witt et al. 2006). This is about 100 times lower than average concentrations at Barbados and elsewhere in the Western Atlantic impacted by African dust (Prospero and Lamb 2003; Prospero 1999). This raises questions about the CALIPSO threshold of detection for dust. An analysis of the depolarization ratio spatial variation may give insight to the dominant aerosol species in the Polluted Dust classification.

In addition, a Polluted Continental Aerosol classification only differs from Smoke in that Smoke is ascribed to elevated layers and Polluted Continental Aerosol to surface layers. Polluted Continental Aerosol frequency of occurrence is generally found at or near regions of greatest Smoke frequency of occurrence. Therefore, it may be beneficial to consider Polluted Continental Aerosol and Smoke in concert for analysis. On the other hand, Polluted Continental Aerosol is often found over densely populated regions in which case it might be linked to pollution emissions. These uncertainties could be resolved by linking CALIPSO attributions to field measurements of aerosol size and composition at altitude and at the surface.

The observed aerosol structures presented here cannot be directly used to assess aerosol–climate interactions. It is our intent that this dataset be used as a validation tool to improve model simulations. Because the aerosol vertical structure is important, the accuracy of simulated aerosol direct and indirect radiative effect will improve by more accurately representing the 3D aerosol structure in the models.

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