

1 CALIPSO observations of transatlantic dust: vertical 2 stratification and effect of clouds

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4 Weidong Yang¹, Alexander Marshak², Tamás Várnai³, Olga V. Kalashnikova⁴
5 and Alexander B. Kostinski⁵

6 [1]{Goddard Earth Sciences Technology and Research, Universities Space Research
7 Association, Columbia, MD 21044 USA}

8 [2]{NASA Goddard Space Flight Center, Greenbelt, MD 20771 USA}

9 [3]{Joint Center for Earth System Technology, University of Maryland at Baltimore County,
10 Baltimore, MD 21228 USA}

11 [4]{Jet Propulsion Laboratory, 4800 Oak Grove Drive, Ms 180-401, Pasadena, CA 91109
12 USA}

13 [5]{Department of Physics, Michigan Technological University, Houghton, MI 49931 USA}

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15 Correspondence to: Weidong Yang (Weidong.Yang@nasa.gov)

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17 **Abstract**

18 We use CALIOP nighttime measurements of lidar backscatter, color and depolarization ratios
19 during the summer of 2007 to study transatlantic dust properties downwind of Saharan
20 sources, and to examine the interaction of clouds and dust. Our analysis suggests that (1)
21 while lidar backscatter doesn't change much with altitude in the Saharan Air Layer (SAL),
22 depolarization and color ratios both increase with altitude in the SAL; (2) lidar backscatter
23 and color ratio increase as dust is transported westward in the SAL; (3) the vertical lapse rate
24 of dust depolarization ratio, introduced here, increases within SAL as plumes move westward;
25 (4) nearby clouds barely affect the backscatter and color ratio of dust volumes within SAL but
26 not so below SAL. Moreover, the presence of nearby clouds tends to decrease the
27 depolarization of dust volumes within SAL. Finally, (5) the odds of CALIOP finding dust
28 below SAL next to clouds are about 2/3 of those far away from clouds. This feature, together
29 with an apparent increase in depolarization ratio near clouds, indicates that particles in some

1 dusty volumes lose asphericity in the humid air near clouds, and cannot be identified by
2 CALIPSO as dust.

3

4 **1 Introduction**

5 Atmospheric mineral dust particles have significant effects on the climate and the
6 environment. Despite notable advances in modeling and satellite and ground-based
7 measurements, dust remains the dominant factor in the uncertainty of aerosol radiative forcing
8 (IPCC, 2001, 2007). Dust emitted from dry areas of Africa is transported over the North
9 Atlantic Ocean to coastal areas of America with a peak of deposition during summer months
10 (e.g., Prospero and Carlson, 1972; Mattsson and Nihlen, 1996; Prospero and Lamb, 2003;
11 Torres et al., 2002; Kaufman et al., 2005).

12 The influence of dust on a radiative budget depends on its ability for absorbing and scattering
13 solar and IR radiation. Dust optical properties are determined by the refractive index (i.e.,
14 chemical composition) (e.g, Sokolik and Toon, 1999; Wang et al., 2002; Lafon et al., 2006;
15 Kahnert et al., 2007; Kandler et al., 2007, 2009; Osborne et al., 2008; Petzold et al., 2009),
16 size and shape of dust particles (Kalashnikova and Sokolik, 2002; Dubovik et al., 2006;
17 Nousiainen, 2009). Dust hygroscopicity describes the particles' ability for taking up water
18 from humid air, and is rooted in the physical-chemical properties of dust components. Dust is
19 mostly composed of water-insoluble minerals and shows nearly complete hydrophobicity or
20 at least poor hygroscopicity (e.g. Twomey, 1977; Li-Jones et al., 1998). However, after being
21 lifted in the air and mixed/coated with water-soluble materials such as sea-salt, sulfate, or
22 nitrate by atmospheric processing (Levin et al., 1996; Yin et al., 2002), dust hygroscopicity
23 can increase, which can cause changes in optical properties. The increase in hygroscopicity
24 may also transforms dust into effective cloud condensation nuclei (CCN) or ice nuclei (IN)
25 (Johnson, 1982; Wurzler et al., 2000; DeMott et al., 2003; Sassen et al., 2003; Lohmann and
26 Feichter, 2005; Twohy et al., 2009a) and thereby affect the formation and distribution of
27 clouds and precipitation (e. g. Kelly et al., 2007), thus also altering the radiative impact of
28 clouds.

29 The evolving shape-dependent optical and hygroscopic properties of dust pose a question:
30 How does the hygroscopicity of dust affect its optical properties near clouds? This question is
31 pertinent as recent studies have shown that optical properties of clear sky aerosols different in
32 the vicinity of clouds from those far away from clouds (e.g., Clarke et al. 2002; Twohy, et al,

1 2002, 2009b; Koren et al., 2007; Su et al., 2008; Redemann et al., 2009; Tackett and
2 Girolamo, 2009; Várnai and Marshak, 2011). The answer to this question is likely not only to
3 improve our understanding of dust-cloud interactions but also yield better estimates of direct
4 radiative forcing. To that end, we present analysis of dust properties over the North Atlantic
5 Ocean—including near-cloud behavior—based on Cloud-Aerosol Lidar with Orthogonal
6 Polarization (CALIOP) data (Winker, et al. 2003).

7 CALIOP is a space based lidar system onboard the Cloud Aerosol Lidar and Infrared
8 Pathfinder Satellite Observations (CALIPSO) satellite launched in 2006. CALIOP data offers
9 many advantages for this study. First, since CALIOP uses a laser with a small footprint (~90
10 m in diameter on the ground), its aerosol data is not affected by the 3D radiative
11 enhancements of nearby clouds, which cause complications for instruments observing
12 reflected sunlight (Wen et al., 2007; Marshak et al., 2008; Várnai and Marshak, 2011).
13 Second, CALIOP provides backscatter depolarization information at 532 nm, which allows
14 one to distinguish (typically) non-spherical dust from (typically) spherical droplets (Sassen,
15 2000; Murayama et al., 2001 Vaughan et al., 2004). Third, CALIOP's high spatial resolution
16 (30 m vertically and 333 m horizontally) is well-suited for studying cloud-dust interactions
17 that have typical scales of several kilometers (e. g., Koren et al., 2007; Várnai and Marshak,
18 2011).

19

20 **2 Data and methodology**

21 This study uses a month-long (06/07/2007-07/07/2007) dataset of nighttime CALIOP Version
22 3 data over the North Atlantic Ocean (0°-45 North, 0°-90° West). We note that since
23 CALIPSO orbits are repeated in a 16-day cycle, our month-long dataset covers almost two
24 orbital cycles, with the longitudes of closest orbits being 1.55° apart. Since in summer there
25 are usually multiple outbreaks per month (Huang et al., 2010), this month-long dataset is
26 sufficient to observe the basic features of dust outbreaks.

27 CALIOP measures the total backscatter of its laser pulses at 532 nm and 1064 nm
28 wavelengths, and the perpendicularly polarized backscatter at 532nm.

29 This paper characterizes dust particles using their attenuated backscatter coefficient (β'),
30 attenuated color ratio (χ'), and depolarization ratio (δ') values. Unless specified otherwise, β'
31 is the median of the vertically averaged 532 nm attenuated backscatter coefficients within

1 dust layers identified by the 5 km-resolution aerosol product. χ' is the ratio between β' values
2 at 1064 nm and 532 nm wavelengths, which tends to increase with the size of particles; this is
3 especially true for optically small spherical particles. δ' is the ratio between the perpendicular
4 and parallel components of β'_{532} , and is greater for non-spherical particles. We note that
5 analyzing measured optical quantities helps avoid potential artifacts that could be introduced
6 by uncertainties in retrieving physical particle properties. However, we still use the results of
7 operational CALIOP data processing for identifying cloud and dust layers. The operational
8 algorithm identifies cloud and dust layers in three steps.

9 First, it identifies particle layers based on the observed 532 nm backscatter values (Winker et
10 al., 2009; Vaughan et al., 2009).

11 Second, it determines whether a detected layer is a cloud or aerosol layer based on its latitude,
12 altitude, 532 nm backscatter, color ratio and depolarization ratio (Liu et al., 2004, 2009). The
13 most obvious clouds are identified at 333 m resolution, while the more ambiguous cases are
14 decided at a coarser (1 km or 5 km) resolution. Our study considers a location cloudy if the 1
15 km resolution Level 2 CALIOP cloud product indicates the presence of clouds. In order to
16 reduce the impact of misclassifications between clouds and aerosols, this paper examines
17 aerosol layers only if the Cloud-Aerosol Discrimination (CAD) product (Liu et al., 2004,
18 2009)—based on probability distribution functions obtained from expert classifications for
19 sample orbits—indicates that the likelihood of aerosol exceeds 70%. As additional precaution,
20 the statistical analysis in this paper examines median values instead of mean values, because
21 medians are less sensitive to any outlying data points influenced by undetected cloud
22 particles. The uncertainty of median values is estimated using the bootstrapping algorithm
23 (Efron and Gong, 1983).

24 Third, the operational CALIOP data processing identifies dust layers as aerosol layers with
25 high depolarization ratio values (Omar et al, 2009). Because depolarization depends on
26 particle shape, it is well suited for separating typically non-spherical dust particles ($\delta' > 0.2$)
27 from usually spherical non-dust aerosols ($\delta' < 0.075$) over ocean. We note that this paper
28 considers dust-containing layers identified as either “dust” or “polluted dust” in the 5 km-
29 resolution Level 2 aerosol product. (Polluted dust is dust mixed with biomass burning
30 aerosols or polluted marine aerosols, with a depolarization ratio between those of dust and
31 non-dust aerosols, 0.075 and 0.2 (Omar et al., 2009)).

1 To discern changes in dust properties during transatlantic transport, we examine dust behavior
2 in the three regions shown in Figure 1: East (E) (0° - 30° W), Middle (M) (30° - 60° W) and
3 West (W) (60° - 90° W). These three regions lie at different distances from the African dust
4 sources, and cover most of the dust paths from Africa to America during the summer of 2007.

5

6 **3 Results and discussion**

7 The spatial and optical characteristics of African dust vary during the transatlantic journey (e.
8 g., Liu et al., 2008; Huang et al., 2010). In this section we examine the variations in three
9 steps. First, the overall statistics of dust properties in the three geographic regions are
10 compared. This part focuses on the vertical distribution of dust samples in the CALIOP 5 km
11 resolution aerosol product, and on the vertical distributions of attenuated backscatter
12 coefficient, color ratio and depolarization ratio. We then analyze the relationships between
13 dust properties and cloud coverage in the three regions. Finally, we discuss the systematic
14 changes in dust properties that occur near clouds.

15 **3.1 Dust properties in the three regions**

16 **3.1.1 Vertical distribution of dust**

17 Figure 2a shows the vertical distribution of dust samples in our three regions, normalized by
18 the total number of dust samples within each region. The number of dust samples is defined
19 as the number of 5 km long 270 m high volumes that, according to the CALIOP aerosol
20 product, contain dust and have a CAD value between -70 and -100. In the Eastern (E) region,
21 more than 80% of dust is between 1.5 km and 5.5 km altitude, with the peak probability
22 around 3.5 km. This elevated dust distribution is a typical result of two confining inversions
23 below and above the SAL (Carlson and Prospero, 1972). The dust remains elevated in the
24 middle (M) region as well, although the mean elevation descends about 0.5 km. The similar
25 patterns in the E and M regions indicate that the driving forces keeping the dust at high
26 altitudes in region E persist in region M as well. If a 3-day average transport time from
27 Region E to M is assumed, the descending velocity from center of E to center of M is
28 estimated around 1.7 mm/s, which is consistent with the typical SAL average descending
29 velocity of 1-2 mm/s (Carlson and Prospero, 1972). Finally, in the West (W) region the
30 chances of finding dust decrease steadily with altitude, and dust is rarely found above 5 km.

1 This dramatic change in the vertical distribution implies that the meteorological conditions in
2 region W are different from those sustaining the elevated profiles in regions E and M.
3 Moreover, the vertical distribution of dust in region W indicates that (dry and wet) dust
4 sedimentation has the strongest impact over region W.

5 **3.1.2 Attenuated backscatter coefficient, color ratio, and depolarization** 6 **ratio of dust**

7 This section examines dust optical properties in the three regions. Since dust above 5 km is
8 rare, the analysis of dust optical properties will be limited to altitudes below 5 km. The
9 analysis uses the Level 2 CALIOP aerosol layer product, which provides averaged β' , χ' , and
10 δ' values for each dust layer. These layer-average values are assigned to all altitude bins
11 within a dust layer when creating Figure 2 (and 4).

12 The results in Figure 2 show that dust properties vary with altitude differently within SAL
13 (from 1.5 km to 5 km in altitude of regions E and M), below the SAL (below 1.5 km in
14 regions E and M), and in region W.

15 In the SAL, median β'_{532} values are nearly constant with altitude, but the medians of χ' and δ'
16 increase with altitude. Since the typically large and non-spherical dust particles imply large χ'
17 and δ' values, the observed dusty volume behavior suggests that the concentration of dust
18 increases with altitude and/or the concentration of non-dust marine aerosols in dusty volumes
19 decreases with altitude.

20 Below the SAL in the E and M regions, the median δ' increases with altitude. This is the
21 result of dust mixing with non-dust marine aerosols in the moist air confined between the
22 marine surface and the inversion created by the dry and warm SAL aloft. Since the
23 concentration of wet marine aerosols is much higher below than inside the SAL, backscatter
24 from these aerosols contributes significantly to the lidar signals and reduce the depolarization
25 ratios of dusty volumes below the SAL. As discussed in the following paragraph, the mixing
26 of dust and marine aerosols in region W could be the major reason behind higher backscatter
27 and lower depolarization in region W than in E or M.

28 In region W, the median of β'_{532} decreases with altitude and the medians of χ' and δ' increase
29 with altitude up to at least 4-5 km. This behavior arises from the dissipation of the SAL in the
30 region. As the SAL extends westward, its temperature drops and convection can bring in

1 moist air from below. Eventually the inversion confining the moist marine air at low altitudes
2 breaks up in region W, which allows the moist marine air to reach much higher altitudes.
3 Even so, the concentration of wetter and larger marine aerosols tends to decrease with
4 altitude, and so their contribution to the lidar backscatter of dusty volumes tends to be smaller
5 at higher altitudes. This results in higher median χ' and δ' values at higher altitudes even in
6 region W.

7 A comparison of dust properties in regions E, M, and W also reveals several features related
8 to SAL influences at different transport stages. For example, at most altitudes the medians of
9 β'_{532} and χ' increase westward, whereas the median of δ' tends to decrease westward. These
10 trends arise from the fading of the SAL during the westward transport: As the air moves from
11 west Africa to the Caribbean, the SAL temperature keeps decreasing, which allows the
12 concentration of moist marine air mixed in from below to keep increasing. The presence of
13 more moist marine aerosols at west increases the median β'_{532} and χ' of dusty volumes and
14 reduces the median δ' value.

15 However, some features in Figure 2 cannot be explained by contributions from non-dust
16 marine aerosols in dusty sample volumes, and are likely caused by changes in the properties
17 of dust particles instead. For example, Figure 2d shows that above 3.5 km, the median δ' is
18 larger in region M than in region E. This cannot be explained by mixing from below, because
19 the larger contribution of mixed-in marine aerosols in region M would imply smaller (rather
20 than larger) δ' values. Instead, the observed tendency is likely related to lower fall speed for
21 aspherical dust particles: as the more spherical particles fall faster, this leaves an increasingly
22 non-spherical dust population at high altitudes as the air moves to region M. The plausibility
23 of this scenario is also supported by simulations for highly irregular particles falling slower
24 than more spherical ones, because of greater air resistance (Ginoux, 2003). This issue will be
25 further discussed in Section 3.3.2.

26 We note that the slight increase of χ' of dusty volumes from E to M (in Figure 2c) is different
27 from the behavior of dust Angstrom exponents retrieved by MISR and MODIS, the latter
28 displaying no significant changes during transatlantic transport (Kalashnikova and Kahn,
29 2008). This contrast is about vertical resolution: the CALIOP trend of westward increase in
30 Figure 2c occurs within SAL but MISR and MODIS integrate the entire atmospheric column
31 and hence are affected by particles above and below SAL.

1 **3.2 Correlation of dust properties with cloud fraction**

2 This section examines the relationships between dust properties and cloudiness in the three
3 study regions. We characterize cloudiness through the cloud fraction (CF), defined for each
4 dust-containing 5 km-size column as the ratio of number of cloudy 0.333 km profiles to the
5 total number of 0.333 km profiles in the column. Simply put, if the number of cloudy 0.333
6 km profiles is m , the cloud fraction is $m/15$. We note that in addition to the cloud fraction
7 varying between 0 and 1, the relative location of dust and clouds within 5 km wide columns
8 can also vary (Figure 3). We also note that unlike the conventional cloud fraction that is based
9 on 2-dimensional (2D) images, our definition here is based on 1-dimensional (1D)
10 measurements along the CALIPSO track. Although off-track clouds may influence dust
11 properties along the track even for $CF_{1D}=0$ (Várnai and Marshak, 2012), CF_{1D} is still a
12 generally useful indicator of cloud coverage.

13 The results in Figure 4 show that dust properties are closely related to CF in all three regions.
14 The main features of the relationship are as follows.

15 First, the top row of Figure 4 reveals that a smaller fraction of dust samples occurs under clear
16 skies in region M than in region E. This is because the SAL is warmer and drier in the East,
17 and so the conditions are less favorable for cloud formation in region E than region M.

18 Second, rows 2 and 3 in Figure 4 reveal that within each region, the median values of β'_{532}
19 and χ' are larger for higher CF s. This feature is likely caused by aerosols getting hydrated and
20 swelling in humid regions containing clouds, although undetected cloud particles may also
21 contribute. The figure also shows that in regions E and M, the increase in backscatter and
22 color ratio is more pronounced below the SAL than inside it. The swelling is greater below
23 the SAL than inside it both because clouds and high humidity are more common below the
24 SAL, and because hygroscopic marine aerosols are fairly abundant at low altitudes even in
25 dust layers, whereas the SAL is dominated by less hygroscopic dust particles.

26 Third, within each region, two opposite trends of correlations appear between δ' and CF :
27 Inside the SAL, the median δ' of dust is always larger in clear sky than in cloudy skies;
28 whereas below the SAL, the median δ' of dust is always smaller in clear sky than in cloudy
29 skies. The domains of these opposite behaviors can be separated in the fourth row of Figure 4
30 roughly at the crossing point of the red curve ($CF=0$) and the blue curve ($0<CF<0.6$). We
31 note that these crossing points are approximately at the altitude of the bottom of the SAL. The

1 opposite trends inside and below the SAL clearly indicate a different dust volume
2 depolarization response to increased humidity. The possible mechanisms affecting the
3 apparent depolarization ratio of dusty volumes below the SAL will be discussed in Section
4 3.4.

5 **3.3 Features of dust volumes in the SAL under clear skies**

6 Unlike the dusty volumes below the SAL, where the dust is mixed with humidified non-dust
7 aerosols, the SAL is dominated by dust particles. This subsection examines several features of
8 dust volumes inside the SAL in regions E and M. To reduce the effects of clouds, dust
9 volumes are limited to only those under clear skies. In addition, region W is excluded because
10 of its low number of dust samples inside the SAL (Figure 2a).

11 **3.3.1 Relationship of depolarization ratio with color ratio and backscatter**

12 Figures 2 and 4 show that while backscatter is fairly uniform vertically, both color ratio and
13 depolarization ratio increase markedly with altitude inside the SAL of regions E and M. These
14 coinciding increases suggest systematic relationship between depolarization ratio on the one
15 hand and color ratio and backscatter on the other (Figure 5). Figure 5a reveals a positive
16 relationship between the depolarization ratio and color ratio of dust volumes in the SAL of
17 regions E and M. In addition, the dynamic ranges of depolarization ratio and color ratio are
18 much wider in region M than in region E. Figure 5b shows a similar relationship between
19 depolarization ratio and backscatter, though with a much smaller dynamic range for
20 backscatter in region E. Figures 5c and 5d confirm that similar relationships are valid for a
21 different dataset (covering 05/25/2008-06/25/2008) as well.

22 The relationships in Figure 5 and the similarity of results from the two independent datasets
23 can be attributed to the steady altitude-dependence of backscatter, color ratio and
24 depolarization ratio inside the SAL. As discussed in Section 3.1.2, these altitude dependences
25 are likely caused by two mechanisms: (i) a decrease with altitude in the concentration of non-
26 dust particles mixed in from below, and (ii) different fall speeds vertically separating the
27 relatively more spherical dust particles from the least spherical ones. This latter mechanism
28 dominates and is further explored in Section 3.3.2.

29 In addition, we note that the depolarization ratio is not only a function of the aspect ratio of
30 particles but is also related to their size (Mishchenko and Hovenier, 1995). Therefore the

1 relationship between δ' and χ' in Figures 5a and 5c may also indicate that backscatter from
2 larger dust particles can be more depolarized.

3 **3.3.2 Relationship between the vertical increase in depolarization ratio** 4 **and longitude**

5 As indicated in Figure 2, the depolarization ratio not only increases with altitude in the SAL,
6 but also has a larger increase rate in region M than E. As mentioned above, the increase may
7 come from more spherical and less spherical dust particles getting vertically separated
8 because of their different sedimentation speeds. The upward increase in δ' could then be
9 stronger in region M simply because the sedimentation process has more time to work by the
10 time the dust reaches region M.

11 Numerous studies have demonstrated that the fall speed of atmospheric particles is related to
12 their shape (e.g., Ginoux, 2003). Since particles with irregular shapes have greater cross-
13 sectional areas and drag-coefficients, they experience stronger drag force in the air—which
14 implies that more irregular particles fall slower than more spherical ones. Note that a sphere is
15 the most compact object (least surface area for a given volume) and it experiences least drag
16 for a given mass. (Here we assume that dust particle shape does not change systematically
17 with particle size, which also greatly impacts fall speed.) As a result, shape-induced vertical
18 separation will ensue as dust is advected westward, with irregular particles increasingly
19 predominant in the upper portions of SAL. At a constant altitude, this stratification is
20 expected to widen the dynamic range of depolarization ratios with downstream distance from
21 the dust source. This is indeed the case.

22 To that end, we divide regions E and M into sub-regions covering 10° wide longitude bands,
23 and examine the average difference between the depolarization ratios at 3 and 4 km altitudes
24 for each region. As shown in Figure 6, the average difference $\delta'_{4 \text{ km}} - \delta'_{3 \text{ km}}$ keeps increasing
25 with the distance from the west coast of Africa. This result implies that the observed change
26 in volume depolarization ratio within SAL is most likely caused by the greater drag of
27 aspherical dust particles.

28 **3.4 Dust volume properties near clouds**

29 Relative humidity usually increases as clouds are approached and this causes nearby aerosols
30 to swell and get hydrated (acquire thin film of water) or even activated as haze (e.g., Twohy et
31 al., 2009b). Observing changes of dust characteristics near clouds can help improve our

1 understanding of the effect of high relative humidity and clouds on dust particles. Figure 4
2 has shown that the backscatter and color ratio of dust volumes increase with cloud fraction
3 both in and below the SAL, whereas the depolarization ratio changes with cloud fraction
4 differently in and below the SAL. This finding indicates that dust properties in and below the
5 SAL are different. This section further examines the near cloud behaviors of dust volumes in
6 and below the SAL. We note that although the base altitude of the SAL may vary during
7 westward transport, as shown in Figures 2 and 4, this analysis uses constant separation
8 altitude of 2 km for convenience. The analysis uses CALIOP Level 1 data to examine changes
9 in backscatter, color ratio, and depolarization as a function of distance to clouds at a
10 resolution of 0.333 km. In this analysis a 0.333 km resolution clear sky profile is considered a
11 dusty profile if it is included in one or multiple 5 km resolution dust layer(s).

12 Figure 7 illustrates the behavior of dust as a function of distance to clouds. The orange curve
13 corresponds to all aerosol samples while the black and green ones to high (in the SAL) and
14 low (below the SAL) dust, respectively. Figure 7a shows that the fraction of dust profiles over
15 all detected aerosol profiles decreases dramatically near clouds for dust at altitudes below the
16 SAL, but remains relatively stable for those in the SAL. The stable behavior in the SAL can
17 be explained by the fact that low clouds confined to the boundary layer by the inversion at the
18 base of the SAL have little impact on humidity inside the SAL. The near-cloud drop in the
19 fraction of dust profiles below the SAL may results from several factors. First, the chances of
20 wet removal are higher near clouds, and this can lower the fraction of dusty profiles. Second,
21 swelling in the humid air near clouds makes particles more spherical (especially if water-
22 soluble particles pollute dust crystals), resulting in the reduction of depolarization ratio thus
23 the dust signature (Omar et al., 2009); consequently, some dust-containing profiles are
24 (mis)classified as non-dust aerosol. Assuming that below 2 km altitude, the fraction of dust
25 profiles is constant beyond 5 km from clouds (as most humidity changes occur within 5 km
26 from clouds), the roughly 2/3 drop in the fraction of dust profiles near clouds implies that at
27 least 2/3 of dust profiles in the MBL are polluted and hygroscopic. This results in CALIOP
28 missing dust in about 1/3 of dust profiles that occur within 5 km from clouds.

29 Figures 7b-7d also show that backscatter, color ratio, and depolarization ratio all increase near
30 clouds for dust layers below 2 km, but they remain fairly constant for dust layers above 2 km.
31 The stable behavior in the SAL occurs because most clouds are below the SAL and have little
32 impact on dust in the SAL. In addition, the dust population in the SAL is dominated by

1 hydrophobic particles. For dust volumes below 2 km, the enhanced backscatter and color ratio
2 may come from the swelling of hygroscopic dust and non-dust particles in the humid air near
3 clouds, or even from cloud contamination. However, the depolarization ratio is expected to
4 decrease and not increase near clouds, as hydrated particles tend to be more spherical than dry
5 particles. Thus the apparent increase in depolarization ratio near clouds for dust volumes
6 below 2 km is somewhat counter-intuitive. A possible explanation is that the hydrated and
7 more spherical dust particles or those heavily mixed with marine aerosols are (mis)classified
8 as non-dust aerosols due to their reduced depolarization ratio; the remaining particle
9 populations will be dominated by hydrophobic dust particles that have irregular shapes and
10 hence higher depolarization ratios.

11 In principle, multiple scattering by undetected cloud fragments could also increase
12 depolarization, but this is likely insignificant, for two reasons: (i) the increase in backscatter is
13 too small to suggest strong multiple scattering near clouds, and (ii) the depolarization ratio of
14 all aerosols (orange curve in Figure 7d) increases only slightly near clouds, which also
15 suggest that cloud contamination should be less significant if any. Another possibility could
16 be that dense dust was misclassified as cloud and the observed trend could come from
17 changes near thick dust, as opposed to near clouds. However, this would increase the fraction
18 of dust profiles near clouds, whereas Figure 7a shows a decrease: If much of the detected
19 clouds were in fact pockets of dense dust, the fraction of dust profiles should increase near
20 them, as dilute dust profiles are more frequent near dense dust than far from it.

21

22 **4 Summary**

23 This paper uses CALIOP lidar data to examine the bulk optical properties of dust layers as
24 Saharan dust moves westward over the Atlantic Ocean. It analyzes dust layers in three regions
25 along the dust transport route, and examines the relationships between dust properties and the
26 amount and proximity of nearby clouds.

27 The study finds that the observed properties of dusty volumes are related not only to the
28 meteorological conditions in the three regions, but also to the speed and duration of dry and
29 wet sedimentation processes. The study examines four characteristics of dust layers: (i) the
30 volume of air containing dust, (ii) lidar backscatter (related to optical thickness), (iii) color
31 ratio (roughly proportional to particle size) at least for spherical particles, and (iv)
32 depolarization ratio (characterizing particle shape, with larger values for irregular particles

1 than for spherical ones). The results show that lidar backscatter and color ratio are smaller,
2 while the depolarization ratio is larger in the warmer and dryer East region.

3 The analysis reveals that the medians of depolarization ratio and color ratio generally increase
4 with altitude in the SAL. The rate of vertical increase in depolarization ratio is significantly
5 larger farther away from Africa's west coast.

6 We find the dusty volume optical properties related to cloud coverage, with backscatter and
7 color ratio increase with cloudiness of surrounding areas. The effects of cloudiness are most
8 prominent for dust below the SAL. The results highlight that sensitivity to cloudiness is very
9 different below and within SAL.

10 The results also reveal other differences between dust volume near-cloud behaviors inside and
11 below the SAL. In the SAL, the fraction of aerosol samples that contain dust doesn't depend
12 on the distance to clouds, neither the median lidar backscatter, color ratio, and depolarization
13 ratio. Below the SAL, the fraction of aerosol samples containing dust decreases near clouds,
14 while the optical properties show noteworthy increases near clouds. The unique features of
15 dust below the SAL indicate that in humid air near clouds only some large dust particles with
16 much higher depolarization ratio are identified as dust by the CALIPSO detection algorithm,
17 and these particles become less frequent near clouds.

18 Earlier studies demonstrated that the degree of irregularity of dust affects dust optical
19 properties and radiative forcing. Our observations further underline the need for assessing the
20 effects of vertical separation in dust depolarization, caused by shape-dependent fall velocity,
21 both in transport modeling and in estimating dust radiative forcing. In addition, our
22 observations of near-cloud behaviors reveal the complexity of dust mixing with other water-
23 soluble aerosols especially in the MBL, and support the hypothesis that dust, or the MBL part
24 of it, becomes hygroscopic through interactions with atmospheric components in moist air,
25 and this significantly affects dust optical properties.

26

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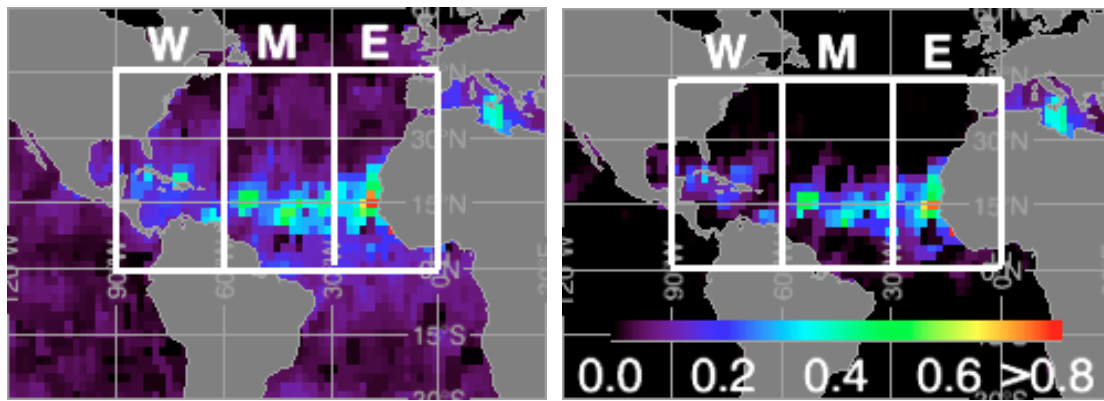
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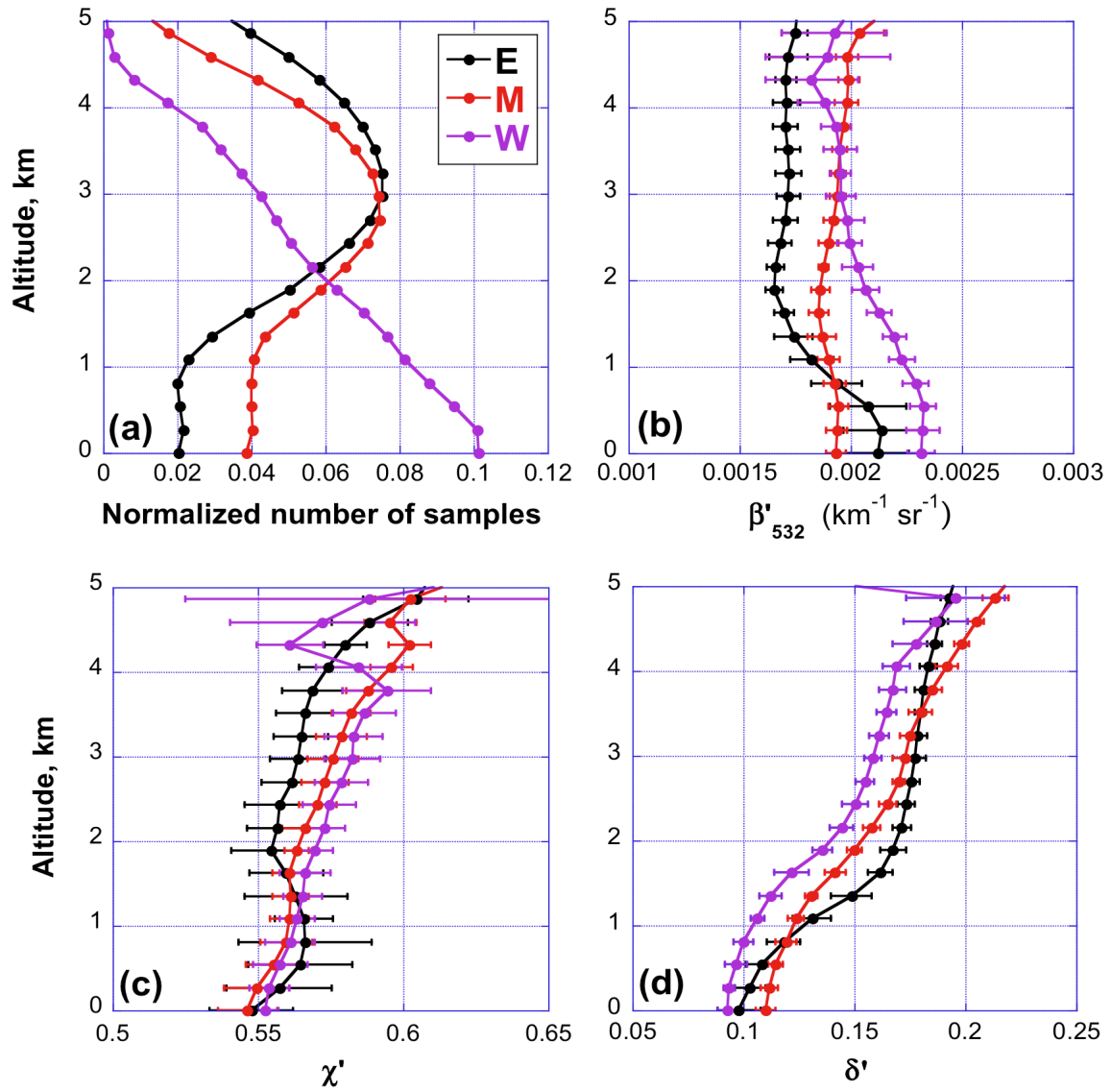


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4 Figure 1. Location of the 3 North-Atlantic regions examined in this study. Colors indicate the
5 nighttime CALIOP Aerosol Optical Depth (AOD) contributed from all aerosols (Left), and
6 contributed from dust aerosols (Right) over oceans for the June 7-July 7, 2007 period, at a
7 pixel resolution of $2^{\circ} \times 2^{\circ}$.

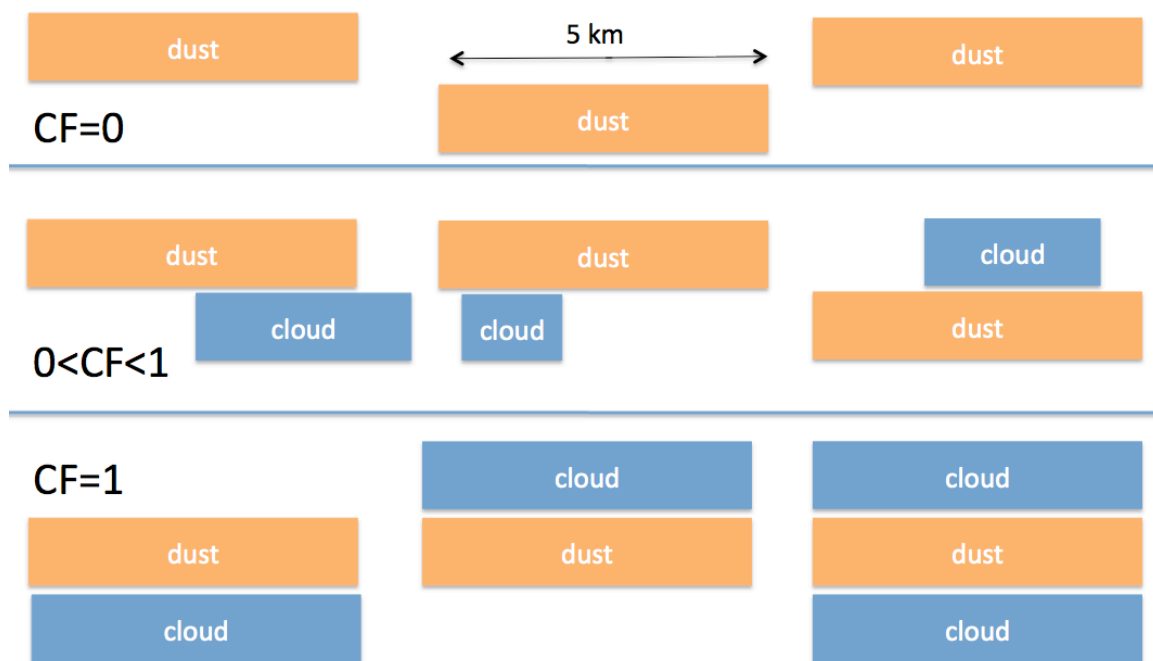
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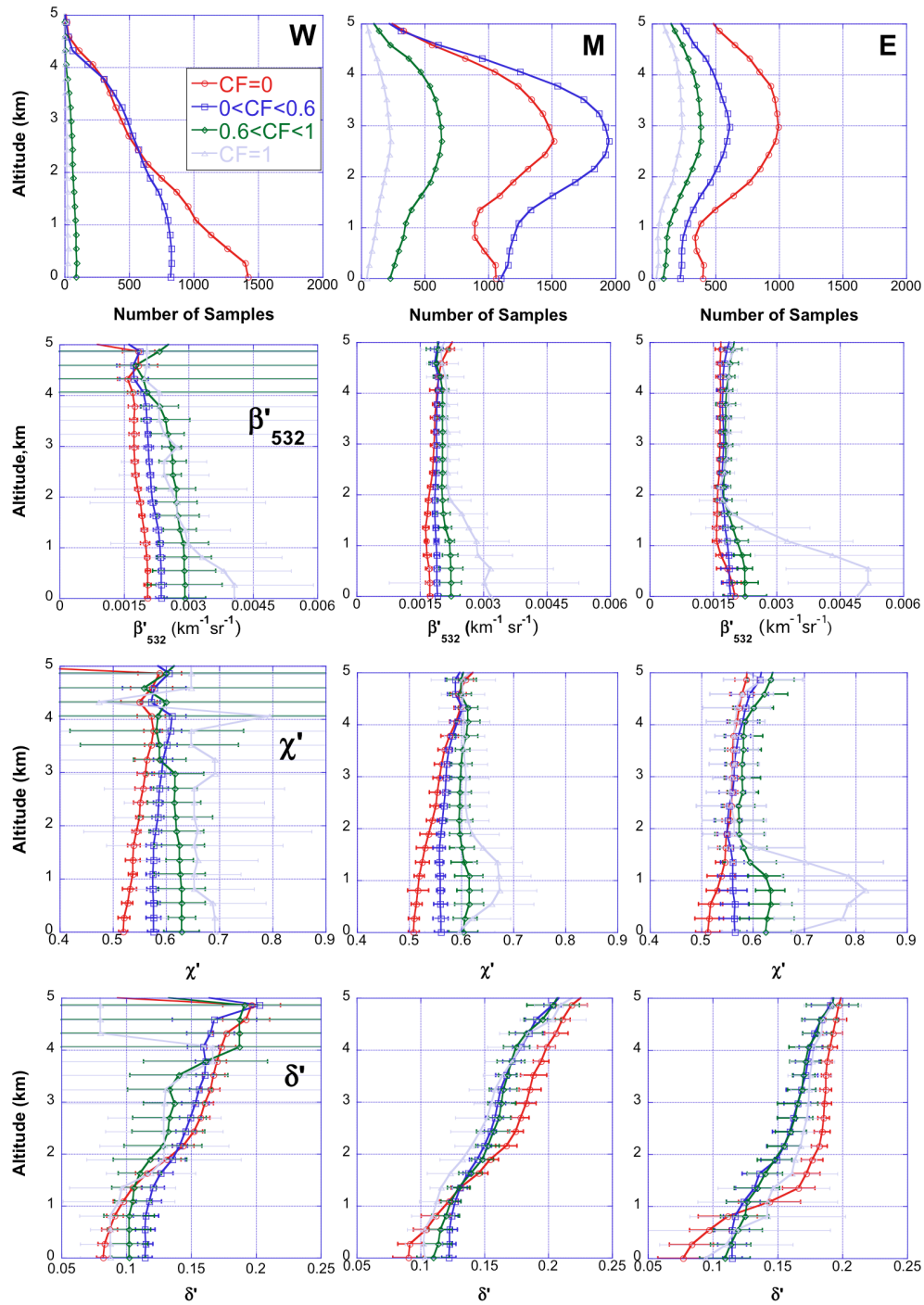
2 Figure 2. Vertical profiles of (a) number of 5 km-resolution dust samples, normalized by the
 3 total number of dust samples at each corresponding region. The total number of dust samples
 4 is 45714 for E, 73141 for M and 32785 for W. In addition, the E, M, and W regions contain
 5 24701, 34019, and 23786 5 km-resolution vertical profiles over ocean, respectively. (b)
 6 median attenuated total backscatter coefficient at 532 nm, β'_{532} , (c) median attenuated color
 7 ratio, χ' , (d) median volume depolarization ratio, δ' . The colors identify the profiles for each
 8 study region (East, Middle, and West). The error bars indicate the uncertainty of median
 9 values, estimated using the bootstrap algorithm.

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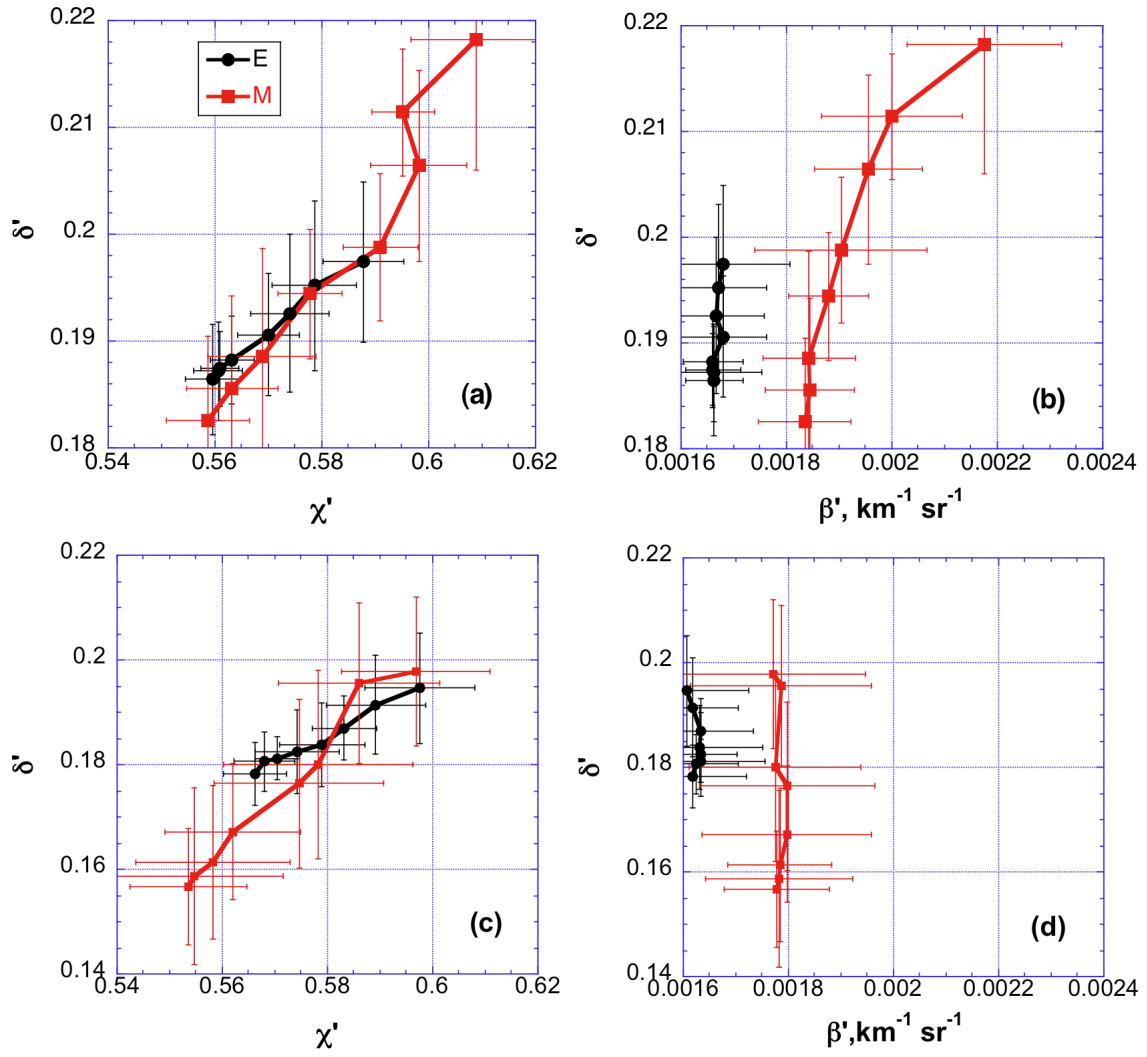
Figure 3. Schematic illustration of Cloud Fraction (CF) definitions for 5 km resolution dust pixels. CF is the fraction of cloudy 0.3 km-resolution pixels in 5 km size areas containing dust.



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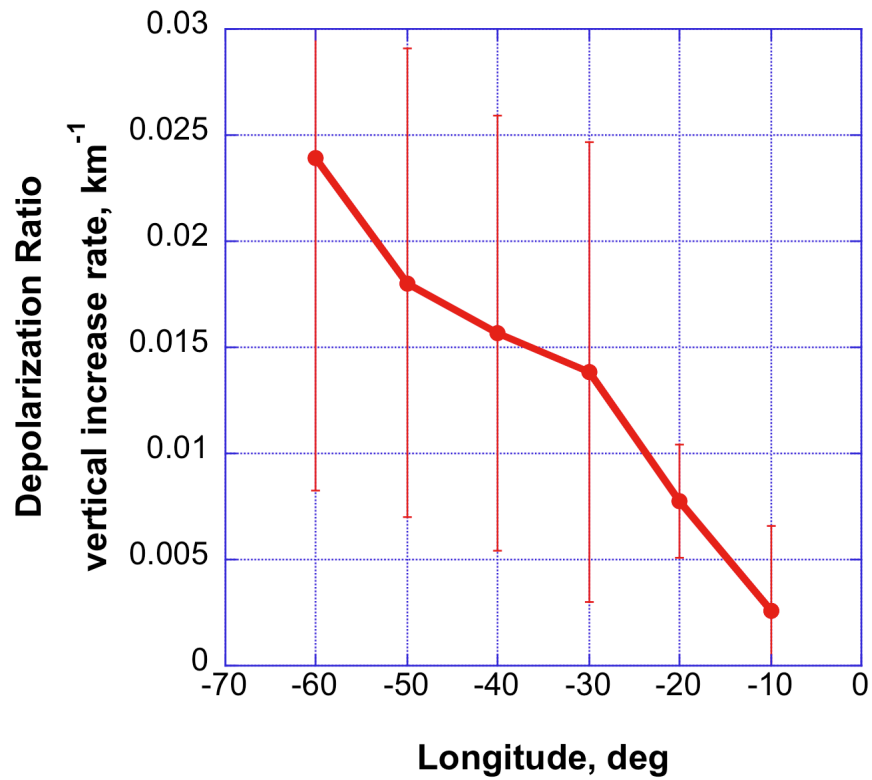
2 Figure 4. Vertical profiles of dust properties for various cloud fractions at the 3 regions. Rows
 3 1, 2, 3 and 4 are for frequency of occurrence, attenuated total backscatter coefficient at 532
 4 nm, β'_{532} , attenuated color ratio, χ' , and volume depolarization ratio, δ' , respectively. The left,
 5 center, and right columns show the West, Middle and East regions, respectively. Results for
 6 different cloud fractions are indicated by different colors.

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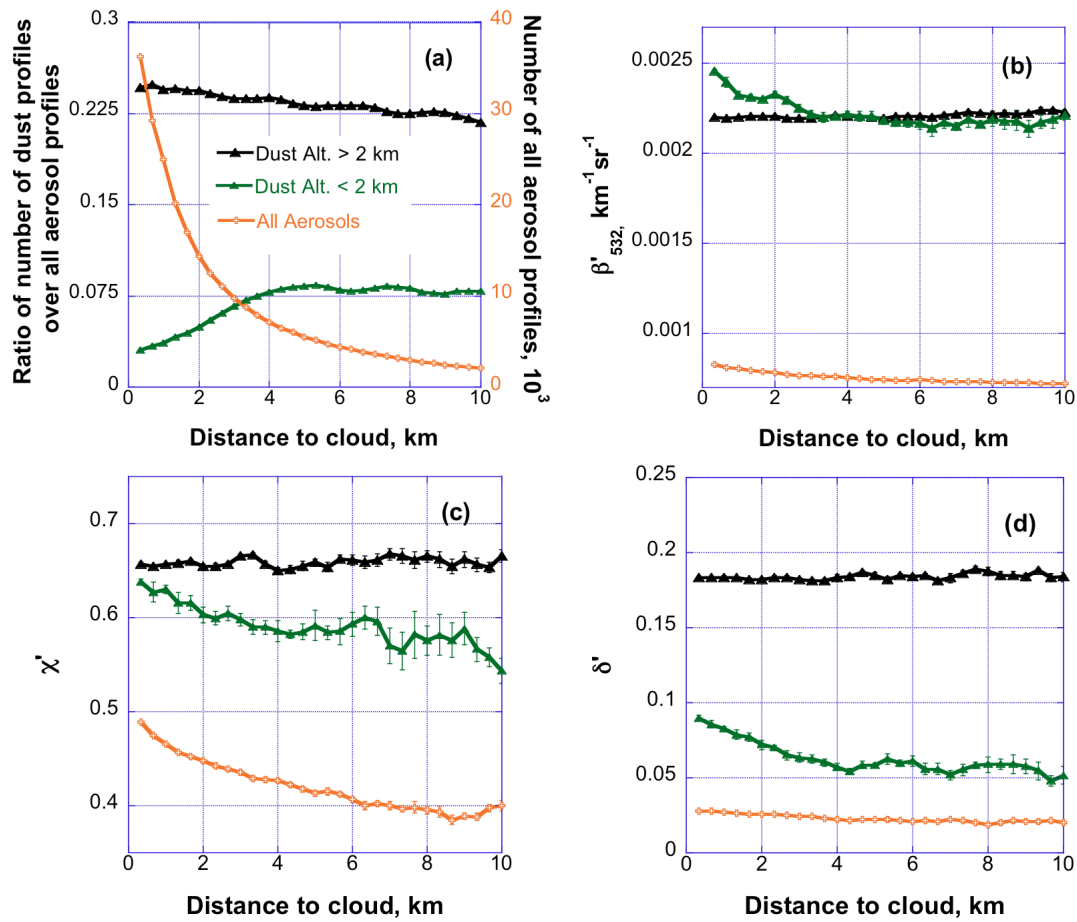
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2 Figure 5. Median depolarization ratio, δ' , as a function of color ratio, χ' (a), and backscatter
 3 coefficient, β'_{532} (b). As in Figure 2, colors identify the examined regions E (black) and M
 4 (red). (c)-(d) are the same as (a) and (b), but for a different dataset from May, 25 to June, 25
 5 of 2008.



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Figure 6. Rate of vertical increase in dust depolarization ratio between 3 km and 4 km altitudes, vs. distance from the African coast, represented by longitude. Particle shape-dependent differences in fall speed cause increasingly more pronounced vertical stratification as plumes move westward.



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Figure 7. Properties of high and low dust as a function of distance to clouds, combined for the three regions (W+M+E): (a) fraction of detected aerosol profiles that contain dust, (b) attenuated backscatter coefficient at 532nm, β'_{532} , (c) attenuated color ratio, χ' , (d) depolarization ratio, δ' . The orange curve in (a) is the number of detected “all aerosol” profiles as a function of distance to clouds. It is also used as denominator in calculating the fraction of high and low dust profiles. Orange curves in (b)-(d) show the optical properties of all aerosols combined.