

# Extinction-to-backscatter Ratios of Saharan Dust Layers Derived from In-Situ Measurements and CALIPSO Overflights during NAMMA

A. H. Omar, Z. Liu, M. Vaughan, K. L. Thornhill, C. Kittaka, S. Ismail, Y. Hu, G. Chen, K. Powell, D. Winker, C. Trepte, E. L. Winstead, B. E Anderson

## Abstract

We determine the extinction-to-backscatter ( $S_a$ ) ratios of dust using (1) airborne in-situ measurements of microphysical properties, (2) modeling studies, and (3) the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) observations recorded during the NASA African Monsoon Multidisciplinary Analyses (NAMMA) field experiment conducted from Sal, Cape Verde during Aug-Sept 2006. Using CALIPSO measurements of the attenuated backscatter of lofted Saharan dust layers, we apply the transmittance technique to estimate dust  $S_a$  ratios at 532 nm and a 2-color method to determine the corresponding 1064 nm  $S_a$ . This method yielded dust  $S_a$  ratios of  $39.8 \pm 1.4$  sr and  $51.8 \pm 3.6$  sr at 532 nm and 1064 nm, respectively. Secondly,  $S_a$  at both wavelengths is independently calculated using size distributions measured aboard the NASA DC-8 and estimates of Saharan dust complex refractive indices applied in a T-Matrix scheme. We found  $S_a$  ratios of  $39.1 \pm 3.5$  sr and  $50.0 \pm 4$  sr at 532 nm and 1064 nm, respectively, using the T-Matrix calculations applied to measured size spectra. Finally, in situ measurements of the total scattering (550 nm) and absorption coefficients (532 nm) are used to generate an extinction profile that is used to constrain the CALIPSO 532 nm extinction profile and thus generate a stratified 532 nm  $S_a$ . This method yielded an  $S_a$  ratio at 532 nm of 35.7 sr in the dust layer and 25 sr in the marine boundary layer consistent with a predominantly seasalt aerosol near the ocean surface. Combinatorial simulations using noisy size spectra and refractive indices were used to estimate the mean and uncertainty (one standard deviation) of these  $S_a$  ratios. These simulations produced a mean ( $\pm$  uncertainty) of  $39.4 (\pm 5.9)$  sr and  $56.5 (\pm 16.5)$  sr at 532 nm and 1064 nm, respectively, corresponding to percent uncertainties of 15% and 29%. These results will provide a measurements-based estimate of the dust  $S_a$  for use in backscatter lidar inversion algorithms such as CALIOP.

## 1. Introduction

Lidar is a powerful tool for studying the vertical distribution of aerosols and clouds in the atmosphere. Of particular importance is the distribution and transport of Saharan dust systems. The deployment of CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations), a joint NASA-CNES satellite mission, has enabled vertically resolved measurements of Sahara air layer(s) (SAL) which will provide significant insights into properties of Sahara dust aerosols. CALIPSO is designed to provide measurements to advance our understanding of the role of aerosols and clouds in the climate system [Winker *et al.*, 2009]. The Cloud-Aerosol Lidar with Orthogonal Polarization [CALIOP, Winker *et al.*, 2007] is the primary instrument on the CALIPSO satellite. CALIOP is designed to acquire vertical profiles of elastic backscatter at two wavelengths (1064 nm and 532 nm) from a near nadir-viewing geometry during both day and night phases of the orbit. In addition to the total backscatter at the two wavelengths, CALIOP also provides profiles of linear depolarization at 532 nm. Accurate aerosol and cloud heights and retrievals of extinction coefficient profiles are derived from the total backscatter measurements [Vaughan *et al.*, 2009]. The depolarization measurements enable the discrimination between ice clouds and water clouds [Hu *et al.*, 2009] and the identification of non-spherical aerosol particles [Liu *et al.*, 2009]. Additional information, such as estimates of particle size for the purpose of discriminating between clouds and aerosols, are obtained from the ratios of the signals obtained at the two wavelengths. On April 28, 2006, the CALIPSO satellite was launched into a low earth sun-synchronous orbit at a 705-km altitude, and an inclination of 98.2 degrees. A few months later, in August 2006, the NASA African Monsoon Multidisciplinary Analyses (NAMMA)

campaign commenced at the Cape Verde Islands, 350 miles off the coast of Senegal in West Africa. NAMMA was designed to study the evolution of precipitating convective systems, largely as this evolution pertained to the SAL and its role in the tropical cyclogenesis. Several aircraft flights were dedicated to nearly coincident measurements with NASA's orbiting satellites (including Aqua, TRMM, and CloudSat/CALIPSO). For this study, we use data collected aboard NASA's DC-8 medium altitude research aircraft outfitted with, among other instruments, a full suite of sensors and probes designed to measure aerosol microphysical and optical properties. Relevant parameters include high spatial-resolution scattering and absorption coefficients at multiple wavelengths in the visible spectrum and dry particle size distributions over the 0.08 to 10  $\mu\text{m}$  diameter range [Chen *et al.*, 2010].

Depending on the mineralogical composition, the SAL can have a significant impact on both the radiation balance and cloud processes. Dust particles scatter in the shortwave regime (cooling the planet) and absorb both shortwave and longwave radiation (heating the planet). By some estimates the anthropogenic forcing due to dust is comparable to the forcing by all other anthropogenic aerosols combined [Sokolik and Toon, 1996]. Saharan dust influences cyclone activity and convection in the region off the west coast of Africa and air quality as far west as the US east coast and Gulf of Mexico. There have been reports of causal links between cyclone activity and dust loading suggesting that perhaps the Sahara dust layer acts to inhibit cyclone development [Dunion and Velden, 2004] and more generally convection [Wong and Dessler, 2005]. Sahara dust is unique in its ability to maintain layer integrity as it is transported over long distances ( $\sim 7500$  km) to the Americas [Liu *et al.*, 2008; Maring *et al.*, 2003; Savoie and Prospero, 1976]. The

77 presence of Sahara dust layers have been found to perturb ice nuclei (IN) concentrations  
78 as far away as in Florida. During CRYSTAL-FACE (Cirrus Regional Study of Tropical  
79 Anvils and Cirrus Layers - Florida Area Cirrus Experiment), DeMott et al. [2003] found  
80 that IN concentrations were significantly enhanced in heterogeneous ice nucleation  
81 regimes warmer than  $-38^{\circ}\text{C}$ , when Saharan dust layers are present. It is therefore  
82 important to study the distribution and optical properties of Sahara dust.

83 In order to estimate the optical depth of the Sahara dust layers from elastic backscatter  
84 lidar measurements, the  $S_a$  ratio must be known or prescribed. Given aerosol free regions  
85 above and below a lofted dust aerosol layer,  $S_a$  can be calculated from the attenuated  
86 backscatter profile of a space-based lidar return [Young, 1995].  $S_a$  for dust aerosols is  
87 dependent on the mineral composition, size distribution, and shape parameters (e.g.,  
88 aspect ratio, and complexity factor). All of these are highly variable and for the most part  
89 not well known. For these reasons,  $S_a$  obtained from scattering models have larger  
90 uncertainties than models of the nearly spherical urban pollution or marine aerosols.

91 There have been several studies and measurements of dust  $S_a$  at 532 nm [Ackermann,  
92 1998; Anderson et al., 2000; Berthier et al., 2006; Di Iorio et al., 2003; Di Iorio et al.,  
93 2009; Muller et al., 2007; Müller et al., 2000; Tesche et al., 2009] and relatively few such  
94 measurements or studies of dust  $S_a$  at 1064 nm [Ackermann, 1998; Liu et al., 2008;  
95 Tesche et al., 2009]. Prior to NAMMA, there were a number of vertically-resolved  
96 measurements of Saharan dust microphysical and optical properties including AMMA  
97 and DODO campaigns. NAMMA studies along with CALIPSO measurements provide a  
98 unique opportunity to compare extinction measurements derived from in situ profile  
99 measurements of total scattering and absorption aboard the NASA DC-8 and CALIPSO

extinction profiles estimated from two wavelength retrieval methods. These profiles by extension provide the constraints from which the lidar ratios can be determined as explained in the following sections. Section 2 discusses the CALIPSO lidar data and its analysis. The NAMMA data and analyses are discussed in Section 3, and coincident CALIPSO-NAMMA measurements are presented in Section 4. In Section 5, the size distributions measured during NAMMA aboard the DC-8 are implemented in a T-Matrix scheme to estimate profiles of  $S_a$  ratios. Section 6 discusses the uncertainty in  $S_a$  using a combinatorial method.

## **2. CALIPSO Lidar Data and Extinction-to-Backscatter Ratio Retrieval Methods**

The CALIPSO lidar data used for these studies are the version 2.01 lidar level 1 attenuated backscatter returns at the 532 nm perpendicular and parallel channels, and 1064 nm total attenuated backscatter. The volume depolarization ratio is determined from the perpendicular and parallel channels and used to identify dust aerosols [Liu *et al.*, 2009; Omar *et al.*, 2009]. For NAMMA underflights of CALIPSO and near spatial coincidences where both missions observed dust layers of optical depths greater than about 0.3, we compare the extinction profiles from in situ measurements to CALIPSO profiles. In such cases, we calculate the extinction using  $S_a$  that was determined using the transmittance method or an  $S_a$  ratio constrained by the in-situ extinction profiles. In both cases, we use the 2-color methods to retrieve the 1064-nm  $S_a$ , after determining the 532-nm  $S_a$ . These two methods, transmittance and 2-color, are discussed below.

## 2.1. Transmittance Methods

The transmittance method uses the following equation describing the relationship between optical depth and integrated attenuated backscatter, as in Platt [1973]:

$$\gamma' = \frac{1}{2\eta S_a} (1 - \exp(-2\eta\tau)) \quad (1)$$

Here  $\gamma'$  is the integrated (from layer base to top) attenuated backscatter,

$$\gamma' = \int_{\text{base}}^{\text{top}} \beta_a(r) T_a^2(r) dr \quad (2)$$

$\tau$  is optical depth,  $\eta$  is a multiple scattering parameter,  $T^2 = \exp(-2\eta\tau)$  is the layer-effective two-way transmittance, and  $S_a = \sigma_a/\beta_a$  where  $\beta_a$  is the aerosol backscatter coefficient and  $\sigma_a$  is the aerosol extinction coefficient. This ratio is assumed constant throughout a feature. Note that the quantities  $S_a$ ,  $\gamma'$ , and  $\tau$  describe characteristics of an aerosol layer, i.e., they are associated with the backscatter and extinction of aerosol particles only. If we define an effective  $S_a$  ratio,  $S^* = \eta S_a$ , we can rewrite Eq. (1) as follows:

$$S^* = \frac{1 - T^2}{2\gamma'} \quad (3)$$

The effective two-way transmittance is typically obtained by fitting the returns both above and below a feature to a reference clear air scattering profile obtained from local

rawinsonde measurements or meteorological model data [Young, 1995]. In this study, the transmittance method is used to determine  $S^*$  from the 532-nm CALIPSO measurements whenever clear air scattering signals are available both above and below an aerosol layer. However, the same method is not applicable to the 1064-nm CALIPSO measurements, because a reliable measurement of the clear air scattering at 1064 nm, which is about 16 times smaller than that at 532 nm, is not available. To determine  $S^*$  at 1064 nm, the 2-color method described in the next subsection is used.

## 2.2. The 2-Color Method

The 2-color or two-wavelength method was first proposed by Sasano and Browell [Sasano and Browell, 1989] and adapted to space-borne lidar measurements using an optimization technique by Vaughan et al. [2004]. The method requires *a priori* knowledge of  $S_a$  at 532 nm and a suitable profile of 532-nm attenuated backscatter amenable to the calculation of 532-nm aerosol backscatter coefficient profiles. For the NAMMA cases described below these preconditions were satisfied. Whenever a suitable region of clear air was identified both above and below an aerosol layer, the  $S_a$  at 532 nm was determined using the transmittance method described above. Here clear air layer is defined as a region of low attenuated scattering ratios with a mean value equal to or less than 1 and a slope with respect to altitude of approximately zero. This is further confirmed by low volume depolarization ratios in a small region ( $\sim 1/2$  km) below the aerosol layer. In cases where coincident NAMMA measurements are available,  $S_a$  ratio at 532 nm is the value that provides the best fit between the retrieved CALIPSO extinction profiles and the NAMMA in situ extinction profiles obtained by summing the total

scattering and absorption measured by a nephelometer and a Particle Soot/Absorption Photometer (PSAP), respectively, aboard the DC-8.

Once  $S_a$  ratio is determined at 532 nm, the value at 1064 nm can be calculated using the 2-color method. We note that this technique can be used to derive  $S_a$  at 532 nm if the value at 1064 nm is known. Given a solution of the particulate backscatter at 532 nm,  $\beta_{532,p}$ , the 2-color method uses a least squares method to minimize the difference between the measured attenuated total backscatter at 1064 nm,  $B_{1064}$ , and the attenuated backscatter at 1064 nm (right hand side of eq. (4)) reconstructed from the extinction and backscatter coefficients at 532 nm.

$$B_{1064}(r) = (\beta_{m,1064}(r) + \beta_{p,1064}) \cdot T_{p,1064}^2(r) \\ = (\beta_{m,1064}(r) + \underline{\chi} \cdot \beta_{p,532}(r)) \cdot \exp(-2 \cdot \underline{S_{1064}} \cdot \underline{\chi} \cdot \gamma_{532}(r)) \quad (4)$$

The only unknowns (underlined) in eq.(4), are the  $S_a$  ratio at 1064 nm,  $S_{1064}$ , and the backscatter color ratio,  $\chi$  (defined as  $\beta_{1064,p}/\beta_{532,p}$ ). These are both intensive aerosol properties defined by the layer composition, size distribution, and shape of its constituent particles. Since these characteristics do not vary substantially in a given aerosol layer, we make the assumption that  $S_{1064}$  and  $\chi$  are constant within the layer. The algorithm details and optimization techniques are discussed at length in Vaughan (2004) and Vaughan et al. [2004]



### 3. Numerical Calculation Based on NAMMA In-situ Measurements

#### 3.1. Aerosol Microphysical Properties

We use measurements of the aerosol size distributions based on number from the Aerodynamic Particle Sizer (APS, TSI Incorporated, Shoreview, MN) and the Ultra-High Sensitivity Aerosol Spectrometer (UHSAS, Droplet Measuring Systems, Boulder, CO). The UHSAS measures the fine mode aerosol size distributions from 0.06 to 0.98  $\mu\text{m}$ , and the APS measures the coarse mode size distributions from 0.6 to 5.5  $\mu\text{m}$ . We use the size distributions to identify the presence of aerosol dust layers. In many cases, these intense dust layers were visually identified by the instrument operators and in some cases specifically targeted by the DC-8 operators for sampling. Additional information about the composition of these layers is available from the NAMMA data archives (<http://namma.msfc.nasa.gov/>). For each size distribution sampled during a 5-second interval we fit the discrete measurements to the best continuous bimodal lognormal size distribution, as shown in Figure 1. The geometric mean radius and standard deviation of a fine and coarse mode derived from the in situ measurements are used in the numerical calculations.

On August 25, 2008, the DC-8 flew through a dense elevated dust layer measuring nearly 1 km in thickness at a mean altitude of about 2.3 km in an hour-long mostly straight and level flight. The APS and UHSAS measured coarse and fine size distributions, respectively, through the dust layer at intervals of 5 seconds. Figure 2 shows the probability distributions of the geometric mean fine and coarse radii of the dust layer. This figure is generated by taking the discrete 5-second size distribution measurements

and fitting these to a bimodal lognormal distribution as described above. For this dust layer, the microphysical properties of mean, median, and standard deviations of the fine (*coarse*) radius distributions, as shown in Figures 2 (a) and (b), are 0.059, 0.061, and 0.0064  $\mu\text{m}$  (0.54, 0.57, and 0.083  $\mu\text{m}$ ), respectively. The mean, median, and standard deviations shown in Figure 2 (c) and (d) for the fine (*coarse*) geometric standard deviation distributions are 1.613, 1.630, and 0.101 (1.495, 1.545, and 0.151), respectively.

The distributions in Figure 2 show that dust properties after lofting of these layers remain relatively unchanged. Other studies [Liu *et al.*, 2008; Maring *et al.*, 2003; Prospero and Carlson, 1971; 1972], have shown the same consistency in properties after long range transport of dust. In each case, the means and medians of the size distribution descriptors are close, i.e., the size descriptors are nearly normally distributed. The standard deviation is a small fraction ( $<0.16$ ) of the means, i.e., the variance of the data is small and thus the layer is quite homogenous with respect to size across the 1 km vertical extent of the dust plume.

### 3.2. Scattering Models

Mie scattering calculations [Mie, 1908], when applied to dust, are adequate for total scattering, albedo and other flux related quantities, but result in large errors when used to retrieve optical depth from satellite reflectance measurements. In particular and central to the theme of this paper, are  $S_a$  ratios calculated from measured size distributions. Mie calculations underestimate  $S_a$  by up to a factor of 2.0 leading to substantial errors in the

lidar derived aerosol optical depths [Kalashnikova and Sokolik, 2002]. This has been known experimentally for quite some time: laboratory measurements by Perry et al [1978] showed non-spherical particles, when compared to spherical particles having the same equivalent volume, enhance side scattering and suppress backscattering. To account for non-sphericity of dust particles, we use T-matrix calculations with the assumption that the dust shapes can be modeled by randomly oriented prolate spheroids. T-Matrix is a matrix formulation of electromagnetic scattering first proposed by *Waterman* [1971], and subsequently improved and extended to much larger sizes and aspect ratios by *Mishchenko et al.* in a series of papers [Mishchenko, 1991; 1993; Mishchenko and Travis, 1994; Mishchenko et al., 1996a; Mishchenko et al., 1996b; Mishchenko and Travis, 1998]. The T-Matrix code used in these calculations is described in detail in *Mishchenko and Travis* [1998]. This method is particularly suitable for light scattering calculations of non-spherical, polydisperse, randomly oriented particles of identical axially symmetric shape with size parameter,  $x$  ( $x = \pi d_p / \lambda$ ;  $d_p$  is particle diameter and  $\lambda$  is the wavelength), smaller than 30.

It is challenging to determine representative statistics of the mean shape for dust particles because of their complexity and variety in shape. These particles are not only confined to desert regions but are ubiquitous in continental areas where they contribute quite significantly to the extinction budget [Omar et al., 1999]. Fortunately, however, randomly oriented prolate and oblate spheroids can adequately represent the scattering properties of non-spherical particles of the same aspect ratio [cf. *Mishchenko et al.*, 1995; *Mishchenko and Travis*, 1994]. The aspect ratio is the ratio of the largest to the smallest

particle dimension. A prolate (oblate) spheroid is a rotationally symmetric ellipsoid with a polar diameter greater (smaller) than the equatorial diameter.

There have been several studies of aspect ratio distributions representative of dust aerosols. From an analysis of scanning electron microscope images of yellow sand particles, Nakajima et al. [1989] found that the distribution of the minor to major particle radius ratio peaked around 0.6, equivalent to an aspect ratio of 1.67. An investigation of mineral dust particle shapes using electron microscopy by Okada et al. [1987] found a mean aspect ratio of 1.4 ranging from 1.0 to 2.3. Hill et al [1984] compared the measured scattering properties of 312 samples of soil dust with the simulated scattering properties of randomly oriented prolate spheroids using T-matrix. They found the distributions of the aspect ratio of prolate spheroids centered near 2.3 most closely reproduced the measured scattering properties. For this study, we use a mean aspect ratio of 2.0 based on the above studies and investigate the sensitivity of the  $S_a$  ratio to aspect ratios ranging from 1.7 to 2.3 partly to account for the dependence of the aspect ratio on size as discussed by Kalashnikova and Sokolik [2004].

There is quite a wide range of estimated and measured mineral dust refractive indices ( $m-ik$ ). For wavelengths of 550 and 1000 nm, d'Almeida et al. [1991] estimate the real part ( $m$ ) of 1.53 and 1.52, respectively, and a spectrally invariant imaginary part ( $k$ ) of 0.008 for dust-like aerosols, and 1.53-i0.0055 and 1.53-i0.001, respectively, for mineral dust. Ackerman (1998) used values of 1.53-i0.0043 and 1.53-i0.0063 to calculate dust  $S_a$  ratios of 19-23 sr and 17-18 sr at 532 and 1064 nm, respectively. These values are much lower

than more recent 532-nm dust  $S_a$  ratios of 40-60 sr [Catrall *et al.*, 2005; Di Iorio *et al.*, 2003; Muller *et al.*, 2007; Murayama *et al.*, 2003; Voss *et al.*, 2001] because the Ackerman study assumed spherical particles.

Retrievals from radiances measured by ground-based Sun-sky scanning radiometers of the Aerosol Robotic Network (AERONET) over a 2-year period yielded dust complex refractive index values of  $1.55 \pm 0.03 - i0.0014 \pm 0.001$  at 670 nm and  $1.55 \pm 0.03 - i0.001 \pm 0.001$  at 1020 nm at Bahrain (Persian Gulf), and  $1.56 \pm 0.03 - i0.0013 \pm 0.001$  at 670 nm and  $1.56 \pm 0.03 - i0.001 \pm 0.001$  at 1020 nm at Solar Village in Saudi Arabia [Dubovik *et al.*, 2002]. Using vertically resolved aerosol size distributions in a scattering model constrained by lidar measurements of aerosol backscattering coefficient at 532 nm, Di Iorio *et al.* (2003) estimated dust refractive indices of 1.52 to 1.58 (real part), and 0.005 to 0.007 (imaginary part). Kalashnikova and Sokolik [2004] calculated effective refractive indices from component mixtures and found values of  $1.61 - i0.0213$  and  $1.59 - i0.0032$  for Saharan dust at wavelengths of 550 nm and 860 nm, respectively. The values for Asian dust, from the same monograph, are  $1.51 - i0.0021$  and  $1.51 - i0.0007$  at 550 nm and 860 nm, respectively. Using a twin angle optical counter, Eidhammer *et al.* (2008) estimated the indices of refraction to be in the range 1.60-1.67 for the real part and 0.009-0.0104 for the imaginary part.

The Saharan Mineral Dust Experiment [SAMUM, Heintzenberg, 2009; Rodhe, 2009] based in Morocco in 2006, produced several independent estimates of the complex refractive indices of Saharan dust. Kandler *et al.* [2009] determined Saharan dust aerosol complex refractive index from chemical/mineralogical composition of  $1.55 - i0.0028$  and

1.57-i0.0037 at 530 nm for small (diameter < 500 nm) and large particles (diameter > 500 nm), respectively. Schladitz et al. [2009] derived mean refractive indices of 1.53 - i0.0041 at 537 nm and 1.53 - i0.0031 at 637 nm from measurements of scattering and absorption coefficients, and particle size distributions. Using similar methods during SAMUM, Petzold et al. [2009] found real parts of the refractive indices of Saharan dust ranging from 1.55 to 1.56 and imaginary parts ranging from 0.0003 to 0.0052. Some of the estimates of refractive indices reported in the literature are summarized in Table 1.

309 Table 1. Summary of complex dust refractive indices from previous studies

Wavelength (nm)	Real Part	Imaginary Part	Source
500	1.50	0.0045	<i>Volz [1973]</i>
1000	1.53	0.008	<i>d'Almeida et al. [1991]</i> , dust-like
550	1.53	0.0055	<i>d'Almeida et al. [1991]</i> , mineral dust
1000	1.53	0.001	<i>d'Almeida et al. [1991]</i> , mineral dust
532	1.53	0.0043	<i>Ackerman [1998]</i> dust
1064	1.53	0.0063	<i>Ackerman [1998]</i> dust
670	$1.55 \pm 0.03$	$0.0014 \pm 0.001$	<i>Dubovik et al. [2002]</i> , Bahrain dust
1020	$1.55 \pm 0.03$	$0.003 \pm 0.001$	<i>Dubovik et al. [2002]</i> , Bahrain dust
670	$1.56 \pm 0.03$	$0.0013 \pm 0.001$	<i>Dubovik et al. [2002]</i> , Solar Village dust
1020	$1.56 \pm 0.03$	$0.001 \pm 0.001$	<i>Dubovik et al. [2002]</i> , Solar Village dust
532	1.52-1.58	0.005-0.007	<i>Di Iorio et al. [2003]</i> , Saharan dust
500	1.42	0.003	<i>Israelevich [2003]</i> , Sede Boker dust
860	1.51	0.0032	<i>Kalashnikova and Sokolik [2004]</i> Saharan dust
550	1.51	0.002	<i>Kalashnikova and Sokolik [2004]</i> Asian dust
860	1.51	0.0007	<i>Kalashnikova and Sokolik [2004]</i> Asian dust
670	1.45	0.0036	<i>Omar et al. [2005]</i> dust cluster
550	1.53	0.0015	<i>Cattrall et al. [2005]</i> mineral dust
1020	1.53	0.0005	<i>Cattrall et al. [2005]</i> mineral dust
300-700	1.60-1.67	0.009-0.0104	<i>Eidehammer et al. [2008]</i> Wyoming dust
537	1.53	0.0041	<i>Schlادitz et al. [2009]</i> Saharan dust
637	1.53	0.0031	<i>Schlادitz et al. [2009]</i> Saharan dust
450	1.55-1.56	0.0003-0.0052	<i>Petzold et al. [2009]</i> Saharan dust
700	1.55-1.56	0.0003-0.0025	<i>Petzold et al. [2009]</i> Saharan dust
530	1.55	0.0028-0.0037	<i>Kandler et al. [2009]</i> Saharan dust

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311 To perform the sensitivity study described in section 6, we use values of the real part of  
312 the refractive index ranging from 1.45 – 1.55 (normally distributed) and the imaginary

part ranging from 0.00067 – 0.006 (log normally distributed) with a central value of 1.50-  
i0.002. For the scattering calculations using NAMMA size distributions, we use the  
central values of the refractive indices along with the nearly instantaneous (5 second  
interval) size distribution measurements to generate profiles of the aerosol properties.  
Figure 3 is a plot of the fine mode, coarse mode, and total phase function of the dust  
plume encountered on August 19, 2006. The fine mode and coarse mode phase functions  
are computed from the mean of the instantaneous size distributions, and the total phase  
function is the area-weighted composite of the fine and coarse mode phase functions. The  
phase functions are driven largely by the coarse mode, especially at 532 nm, and exhibit a  
more pronounced peak in the backscattering direction at 532 nm than 1064 nm

#### **4. Data Analyses of CALIPSO NAMMA Coincident Measurements**

For this study we analyzed coincident measurements of the CALIOP 532-nm extinction  
profiles and in situ extinction profiles measured at wavelengths near the CALIOP green  
channel. The in situ extinction coefficient is obtained by summing the scattering (550  
nm) and absorption (532 nm) coefficients measured by a nephelometer and a PSAP,  
respectively. Data from the nephelometer and PSAP have been corrected for errors  
associated with the limited detector viewing angle [*Anderson and Ogren*, 1998] and  
scattering from the filter media [*Virkkula et al.*, 2005], respectively. We make the  
assumption that the scattering properties are invariant over the 532 – 550 nm range for  
these large dust particle sizes. We chose three days on which there were near collocated  
CALIPSO and NAMMA measurements of nearly the same airmass.



#### 4.1. August 19, 2006 NAMMA Flight 4

August 19 was one of the days the DC-8 performed an under flight of CALIPSO. Figure 4 shows the time-altitude flight track of the DC-8. A nearly coincident in-situ profile was obtained during the second ascent leg shown in the figure. Atmospheric context for this flight is given by Figure 5(a) and (b), in which both the DC-8 and CALIPSO flight tracks are superimposed on images of measurements made by the Moderate Resolution Imaging Spectroradiometer (MODIS) and Measurements of Pollution in the Troposphere (MOPITT), respectively. The DC-8 flight tracks are the black irregular octagons and the CALIPSO orbit tracks are the straight lines in both images. The underflights in the figures are segments where the DC-8 flight tracks are nearly exactly collocated and parallel to the CALIPSO flight tracks. The MODIS optical depth near the coincident flight segment is about 0.5. The MOPITT image shows moderate CO concentrations in the vicinity of the coincident flight track and therefore indicates that most of the aerosol is dust and not continental pollution or biomass burning. This can also be confirmed by the CALIPSO depolarization measurements. The image also shows high CO concentrations ( $>2.5 \times 10^{18}$  molecules/cm<sup>2</sup>) to the south of the DC-8 flight tracks most likely due to biomass burning, and identified as such by the CALIPSO aerosol subtyping scheme illustrated later in this section.

Unfortunately, the direct underflight of CALIPSO by the DC-8 was a nearly level flight with no in situ profile information. In fact the DC-8 was at a high altitude near Flight Level 330 (~ 10 km) throughout the underflight and therefore did not encounter any significant aerosol layer at this altitude to sample. We used the ascending leg of the DC-8

flight which corresponds to the flight segment during ascent to the CALIPSO underflight portion denoted by blue dots in Fig 5a. The in situ profiles segment is shown in Fig. 4.

The CALIPSO browse images (e.g., Fig. 6) are plots of the attenuated backscatter color coded by intensity varying from blue (weak) to white (very strong). A horizontal line near the 0 km mark denotes the surface. The CALIPSO data used for comparison with the in-situ data come from the 80 profiles (~27 km horizontal average) between the white lines in the browse image (Fig. 6). To determine the optimal 532-nm  $S_a$  ratio for these data, we iteratively adjust  $S_a$  until the difference, in a least squares sense, between the retrieved CALIPSO profile,  $\sigma_{a,532\text{-nm}}$ , and the measured NAMMA profile,  $\sigma_{a,550\text{-nm}}$ , is minimized.

Figure 7 is a plot of the extinction profiles retrieved from CALIPSO's 532-nm backscatter profiles and the profiles of the sum of scattering (550 nm) and absorption (532 nm) measured by the nephelometer and the PSAP aboard the NASA DC-8, respectively. These profiles were taken during the ascent leg shown in Figure 4 corresponding to the flight tracks shown in Figure 5(a). The boundary between the two layers is determined by the increase in the extinction coefficient near 2 km. The two  $S_a$  ratios are values that provide the best fit in a least squares sense of the CALIPSO data to the NAMMA in-situ measurements. The 532-nm  $S_a$  ratios are consistent with a dust plume ( $S_a = 35.7$  sr) above seasalt in the marine boundary layer ( $S_a = 25$  sr). The root mean square (rms) of the differences between the CALIPSO and the NAMMA in-situ extinction coefficient profiles are  $20.2 \text{ Mm}^{-1}$ , and  $47.4 \text{ Mm}^{-1}$ , for the marine and dust layers, respectively. The rms of the differences in the extinction coefficients in the clear

region (4 to 8.5 km) above the dust layer in Fig. 6 is  $12.3 \text{ Mm}^{-1}$ . We see some differences in the two extinction profiles in Figure 7 likely due to the temporal and spatial differences of 30 minutes and 160 km, respectively, between DC-8 and CALIPSO.

Figure 8 is an image of the aerosol scattering ratios measured by the Lidar Atmospheric Sensing Experiment (LASE) on board the DC-8 during the ascent leg of Flight 4 on August 19, 2006 as indicated in Figure 4. The measurements were made at 815 nm and the extinction calculation was performed using an  $S_a$  ratio of 36 sr [Ismail *et al.*, 2010]. The DC-8 flight track is shown by the solid line in Figure 8 during which the in-situ measurements shown in Figure 7 were made. The figure also shows the region nearest to the CALIPSO overpass where measurements of the CALIPSO profiles shown in Figure 7 were made. LASE also observed a dust layer extending to an altitude of 6 km at the coincident point. This dust layer is optically and geometrically thick, and is lofted over a layer of lower optical depth (aerosol scattering ratio  $\sim 1$ ) in the marine boundary layer. Some differences in altitude and extent of the layer between the CALIPSO and LASE measurements can be attributed to temporal and spatial mismatch.

Figure 9 is a plot of the results of (a) the cloud-aerosol discrimination and (b) aerosol classification algorithms applied to the data shown in the browse image including the NAMMA underflight (Flight 4 of August 19, 2006). The CALIPSO level 2 algorithms first discriminate between aerosol and clouds [Liu *et al.*, 2009] and then classify the aerosol layers into aerosol subtypes [Omar *et al.*, 2009]. Figure 9(a) shows that some of the optically thick aerosol near  $15^\circ \text{ N}$  was misclassified as clouds, and thus was not

examined by the aerosol subtyping algorithm. The presence of biomass burning smoke and polluted dust (mixture of dust and smoke) during the first part of the flight depicted in Figure 9(b) is borne out by the high CO concentrations in the MOPPITT data to the south of the DC-8 flight tracks shown in Figure 5(b). Though the small lump of aerosol at the surface near 15° N is classified as pure dust, it is more likely a mixture of dust and marine aerosol. The white line in the Figure is the midpoint of the 80 profiles averaged for the retrieval of the extinction profile discussed above.

CALIPSO also measured a dense Saharan dust layer to the southwest of the coincident measurements during a nighttime orbit on the same day (August 19, 2006). The browse images of attenuated backscatter at 532 nm for this measurement are shown in Figure 10. The inset map in Figure 10 shows the CALIPSO ground track in blue. This dust layer appears to be a more robust part of the same dust plume observed during the coincident measurement.

As shown (by the red dotted oval) in Figure 10(a), the layer exceeds 1000 km in horizontal extent (from 18.3N to 10.3N). The 532-nm aerosol optical depth (AOD) is greater than about 0.3 across most of the 1000-km orbital segment shown in Figure 10(a). Figure 10(b) is a magnified illustration of the region in 9(a) subtended by the yellow dotted line. We divided the layer into five segments and applied the transmittance method of section 2.1 to calculate a 532-nm  $S_a$  and the 2-color method of section 2.2 to calculate a 1064-nm  $S_a$  ratio. These values are shown in green (532 nm) and red (1064 nm) in Figure 10(b). Clear air regions above and below each dust layer were identified

manually, by inspection of the profiles. Note that for this mesoscale layer the 532-nm  $S_a$  ratios range from 38 to 41 sr with an average of 40.1 sr and the 1064-nm  $S_a$  ratios range from 45.8 to 54.2 with an average of 50.9 sr.

Figure 10 shows that the Sahara dust layers once elevated are consistent both geometrically (the layer is confined to 3-5 km altitude band) and optically ( $S_a$  variation at both wavelengths is small and the layer optical depth is near 0.3). The  $S_a$  ratio is an intensive aerosol property that depends on the composition, size distributions, and particle shape of the aerosol and its consistency is an indication that these layers stay intact over very long distances. Other studies have shown the transport of relatively unmixed Saharan mineral dust to the south American rainforest [Ansmann *et al.*, 2009; Graham *et al.*, 2003] and western Atlantic Ocean [Formenti *et al.*, 2003; Kaufman *et al.*, 2005], including the US eastern seaboard (cf. Liu *et al.* 2008).

#### **4.2. August 26, 2006**

The August 26 DC-8 flight included an underflight of CALIPSO. As is the case with the coincident CALIPSO-NAMMA measurements on August 19, the collocated measurements are at one level and lack in-situ profile measurements. In situ profiles of the size distributions were estimated from the DC-8 data obtained during the descent flight segment shown by a red dashed tilted oval in Figure 11 (a) and flown about two hours earlier than the CALIPSO underflight. The MOPITT CO levels (Figure 11b) for this period are slightly elevated. The extinction comparison shows that the layer observed by CALIPSO is more elevated than the one encountered by the DC-8. Figure 12 is a browse image of the CALIPSO 532-nm attenuated backscatter coefficients measured

during the orbital segment corresponding to Flight 8 of the DC-8. The CALIPSO data used for comparison with the in-situ extinction profiles were extracted from the region between the two white lines shown in the figure.

Figure 13 compares the CALIPSO extinction profile with a profile of the extinction derived from in situ measurements (i.e., the sum of the absorption and scattering coefficients). The maximum extinction coefficients ( $160$  and  $140 \text{ Mm}^{-1}$  by CALIPSO and the DC-8, respectively) are comparable, showing that the layer is intact after two hours. The altitudes of the maximum layer extinctions for the CALIPSO and NAMMA measurements are offset by at least one kilometer. The in situ measurements and CALIPSO observations are far removed from each other in this case (two hours and  $1250 \text{ km}$ ). The data shown in Figure 12 is north of the NAMMA flight segment. These differences are shown by the mismatch in layer heights shown in Figure 13. Because of the relatively large mismatch, the constraint method, which requires good coincidence, is not applicable to this case to derive  $S_a$ . However, the transmittance technique can be applied using the CALIPSO measurement averaged over the region bounded by two white lines in Figure 12. Note that the dust layer overlies a streak of marine stratus just above the marine boundary layer. At  $532 \text{ nm}$ , the dust layer has an optical thickness of  $0.41$  and an  $S_a$  ratio of  $38.2 \text{ sr}$ . Though the Saharan layers can have horizontal extents of thousands of  $\text{km}$ , it is possible that the layer observed by CALIPSO is not the same as the one measured by the in situ instruments aboard the DC-8. Notwithstanding this possibility, the properties measured by the two methods provide independent characterization of the Saharan dust layer(s).

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474 **4.3. September 1, 2006**

475 Flight 10 of the DC-8 on September 1, 2006 neither directly underflew nor intercepted  
476 the CALIPSO orbit tracks. However, this flight sampled some of the same aerosol layers  
477 measured during a CALIPSO orbit track, as shown in the MODIS aerosol optical depth  
478 image in Figure 14(a). The region of interest is denoted by the red circle. The DC-8 flight  
479 segment most relevant and of closest proximity is the descending leg located in this  
480 region. The MOPITT CO concentrations are low ( $1.5 - 2 \times 10^{18}$  molecules/cm<sup>2</sup>) in the  
481 comparison region. It is therefore likely that most of the aerosol mass is Saharan dust.  
482 CALIPSO preceded the DC-8 by 14 hours. As in the previous flight of August 26, the  
483 observed layers by CALIPSO and the in situ measurements may be different.

484

485 The CALIPSO browse image (Figure 15) shows the layers of interest marked by two  
486 white lines and comprising of 80 profiles. The MOPITT image of this area shows no  
487 enhancement of CO in the sampling region subtended by a red circle in Figure 14(a). The  
488 transmittance and 2-color methods yielded  $S_a$  ratios of 39.8 sr and 56 sr at 532 nm and  
489 1064 nm, respectively. The optical depth determined from the inversion of the CALIOP  
490 attenuated backscatter using this lidar ratio (39.8 sr at 532 nm) is 0.55. In Figure 14(a),  
491 the MODIS AOD is about 0.5 in this region.

492

493 It is a general transport pattern that Saharan dust aerosol goes through a phase of rising  
494 motion near the source then relative horizontal transport culminating in descent and  
495 deposition near the Americas [Ansmann *et al.*, 2009; Formenti *et al.*, 2003; Graham *et*

*al.*, 2003; *Kaufman et al.*, 2005; *Okin et al.*, 2004]. If the layer observed on September 1, 2006 by CALIPSO is the same as the one observed by the in situ measurements, then it was in the rising phase at a rate of approximately 0.1 kilometers per hour. The rate of ascent is based on the time difference between the CALIPSO overpass (3:30 am Local Time) and DC-8 flight track (6:30 pm Local Time) of nearest approach. The extinction profiles' comparison (Figure 16) between the DC-8 in-situ measurements and the CALIPSO measurements shows an offset of 1.5 km in the altitude of maximum extinction coefficient. The layer shown at the same location has been elevated by 1.5 km since CALIPSO sampled it. During this rising phase, there is no evidence of significant deposition, since the maximum extinction coefficient, a property of the aerosol loading, does not decay appreciably.

## **5. Extinction-to-backscatter ( $S_a$ ) ratios calculation based on NAMMA in situ measurements**

To determine profiles of  $S_a$  ratios and validate the retrieved values, we perform numerical calculations for Saharan dust based on NAMMA in-situ size distribution measurements. We use the DC-8 APS and UHSAS measurements (Chen et al., 2010) to determine coarse and fine size distributions, and then calculate coarse and fine mode phase functions, as in Figure 3, using a T-Matrix scheme. The  $S_a$  ratio of the aerosol is derived from an area-weighted integral of the two modes. Figure 17 is a plot of the profile of  $S_a$  ratios of the 2-km dust layer encountered by NAMMA Flight 4 on August 19, 2006. The figure shows a profile of the coarse number concentration which marks the bottom and top of the layer at 2.5 and 4.6 km, respectively. This is very similar to the coincident CALIPSO extinction



profile shown in Figure 7. The 532-nm and 1064-nm  $S_a$  ratios calculated by this method are  $34.3 \pm 2.0$  sr and  $50.2 \pm 5.7$  sr, respectively. These are in good agreement with  $S_a$  ratios of 38 to 41 sr at 532 nm and 45.8 to 54.2 sr at 1064 nm independently determined from CALIPSO measurements using the transmittance technique (Figure 10) on the same day, albeit further downfield.

Part of the flight on August 25, 2006 was dedicated to an intercomparison of the in situ measurements on the NASA DC-8 and the British BAe146. The DC-8 made a nearly straight and level flight through a dust cloud near 2 km. Figure 18 (a) and (b) show the DC-8 altitudinal flight tracks, and the calculated  $S_a$  ratios (and the coarse number concentration) for this flight, respectively. The dust layer is quite tenuous with maximum coarse number concentrations  $\sim 20 \text{ cm}^{-3}$ . Scattering coefficients varied from 50 to  $75 \text{ Mm}^{-1}$  on intercomparison legs near 19 deg N latitude. Condensation Nuclei (CN) concentrations in the dust layers were fairly low, around  $300 \text{ cm}^{-3}$ , while CO mixing ratios were  $\sim 85$  ppbv and RH was  $\sim 50$  to 60%. Both the CCN and CO concentrations infer the predominance of dust in the aerosol layer. The profiles of the 532- and 1064-nm  $S_a$  ratios shown in Figure 18 for this dust layer are quite consistent with means of  $38.0 \pm 2.5$  sr, and  $48.7 \pm 3.2$  sr, respectively. The small standard deviations in both  $S_a$  ratios indicate that the layer remains very uniform with respect to this optical property.

Size distribution measurements were made during the NAMMA DC-8 Flight 8 on August 26, 2006 of a low density dust layer between 0.6 and 1.6 km. Profiles of  $S_a$  ratios calculated from these measurements are shown in Figure 19. The calculated values are  $39.0 \pm 1.5$  sr and  $45.9 \pm 2.2$  sr at 532-, and 1064-nm, respectively.

This layer is optically thinner than the dust layer encountered on August 19, 2006. Nevertheless, the  $S_a$  ratios determined by T-Matrix calculation for this layer and the direct measurement for the denser layer on August 19, 2006 (40.1 and 50.9 sr at 532 and 1064 nm, respectively) are quite close. The calculated  $S_a$  ratio at 532 nm is also consistent with the value (38.2 sr) retrieved from the CALIPSO measurements on the same day.

During the return flight on August 26, 2006, the DC-8 performed a stair-step descent flight consisting of two level sections and two descent sections. The first straight and level section (Leg 1 in Figure 20a.) was flown at an altitude of  $\sim 2.25$  km in a dense dust plume with coarse number concentrations near  $35 \text{ particles cm}^{-3}$ . The mean  $S_a$  ratios are  $42.4 \pm 1.3$  and  $53.3 \pm 2.0$  sr at 532 nm and 1064 nm, respectively. The plume is fairly consistent as shown by the small standard deviations in the  $S_a$  ratios at both wavelengths. Fig. 20(b) are profile plots of the two descent legs (leg 2 and 4 shown in plot (a) the flight path). The break in ordinate demarcates the straight and level section (leg 3) of the flight. The 532 nm and 1064 nm  $S_a$  ratios for leg 2 are  $46.6 \pm 1.3$  sr, and  $52.6 \pm 1.7$  sr, respectively. In leg 4, the aircraft has begun its descent into the marine boundary layer and there is a sharp decline in the coarse number concentration. The  $S_a$  ratio at 532 nm has also dropped considerably signifying a change in the aerosol composition. The calculated  $S_a$  ratios for leg 4 are  $32.8 \pm 1.5$  sr, and  $51.4 \pm 10.8$  sr, at 532 and 1064 nm, respectively. Note that for leg 4, the 532 nm  $S_a$  ratios are fairly consistent while the 1064 nm values are quite noisy and decrease at lower altitudes. The decreasing trend in lidar ratios at 1064 nm with altitude corresponds to a decrease in the aerosol coarse mode number concentration.

Figure 21 shows the flight path for the in-situ measurements on August 20, 2006. The time is in seconds after midnight UTC. Plot (b) shows a profile plot of the 532 nm and 1064 nm  $S_a$  ratios observed during the descent phase through a dust layer extending from an altitude of 1.6 km to 4.8 km. The 532 nm and 1064 nm  $S_a$  ratios for this dust layer are  $40.8 \pm 3.2$  sr, and  $51.6 \pm 3.8$  sr, respectively. Plot (c) shows the profiles of the 532 nm and 1064 nm  $S_a$  ratios observed during the ascent phase through the dust layer. The 532-nm and 1064-nm  $S_a$  ratios for this dust layer are  $42.8 \pm 3.0$  sr, and  $51.8 \pm 3.4$  sr, respectively. The similarity of the vertical extent and the  $S_a$  ratios at each wavelength suggests that the same dust layer was sampled during both the ascent and descent legs of the flight. As noted before, these layers have spatially and temporally uniform  $S_a$  ratios and perhaps by inference, fairly constant compositions, and are geometrically quite stable.

Figure 22 is a histogram of all the 532-nm and 1064-nm  $S_a$  ratios (~1100 points) determined using the size distributions measured during NAMMA for this study. There is very little overlap of the two nearly normally distributed  $S_a$  ratios. The 532-and 1064-nm mean  $S_a$  ratios ( $\pm$  one standard deviation) are  $39.1 \pm 3.5$  sr and  $50.0 \pm 4.0$  sr, respectively. The 532-nm values ranged from 30 to 53 sr and the 1064-nm values ranged from 32 to 66 sr, in both cases within the estimated ranges of 10 – 110 sr for all aerosol types [Ackermann, 1998; Anderson *et al.*, 2000; Barnaba and Gobbi, 2004]. Figure 23 is a plot of the frequency distribution of the ratio of  $S_a$  ratios, i.e.,  $S_a(1064 \text{ nm}) / S_a(532 \text{ nm})$ , a parameter used in the lidar ratio determination scheme outlined in Catrall *et al.* (2005). The plot shows that for the Sahara dust sampled during NAMMA there is very little

spread in the ratio of  $S_a$  values. The mean 1064 nm  $S_a$  ratio is about 30% larger than the mean 532 nm value for this Saharan dust with a standard deviation of 10%, i.e.,  $S_a(1064 \text{ nm}) / S_a(532 \text{ nm}) = 1.3 \pm 0.13$ . Since the  $S_a$  ratio is an intensive property of the aerosol, its ratio is also an intensive property. The small spread in the ratios of  $S_a$  denotes that the Saharan dust aerosol observed during this period is quite consistent at least in its size distributions. To explore the effects of varying composition (refractive indices) and shape (aspect ratios) on the T-Matrix calculations, both of which were not directly measured for this study, we use the uncertainty analysis described in the next section. Table 2 summarizes the  $S_a$  ratios obtained for Saharan dust aerosols during NAMMA using the three methods. The 532-nm values are fairly consistent while there is a somewhat wider spread in the 1064-nm values. This is particularly interesting because these measurements were made on various days and at various locations.

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Table 2. Summary of  $S_a$  ratio measurements and calculations

	$S_a$ (532 nm)	$S_a$ (1064 nm)
	sr	sr
Constrained method		
08/19/2006	35.7	NA
Transmittance + 2-color methods		
08/19/2006	38.9	53.5
08/19/2006	41.4	50.9
08/19/2006	38.1	54.2
08/19/2006	41.0	45.8
08/19/2006	41.0	50.2
08/26/2006	38.2	NA
09/01/2006	39.8	56.0
Mean	$39.8 \pm 1.4$	$51.8 \pm 3.6$
T-Matrix using NAMMA size distribution measurements		
08/19/2006	$34.3 \pm 2.0$	$50.2 \pm 5.7$
08/25/2006	$38.0 \pm 2.5$	$48.7 \pm 3.2$
08/26/2006	$39.0 \pm 1.5$	$45.9 \pm 2.2$
08/26/2006	$42.4 \pm 1.3$	$53.3 \pm 2.0$
08/26/2006	$46.6 \pm 1.3$	$52.6 \pm 1.7$
08/26/2006	$32.8 \pm 1.5$	$51.4 \pm 10.8$
08/20/2006	$40.8 \pm 3.2$	$51.8 \pm 3.4$
08/20/2006	$42.8 \pm 3.0$	$51.8 \pm 3.4$
Mean	$39.1 \pm 3.5$	$50.0 \pm 4.0$
Noise Laden T- Matrix Simulation		
Mean	$39.4 \pm 5.9$	$56.5 \pm 16.5$

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## 6. Uncertainty Analysis of the modeled $S_a$ ratios

In this section we attempt to propagate the uncertainty in the input variables to the calculated  $S_a$  ratio and determine the overall uncertainty in the calculated dust 532-nm and 1064-nm  $S_a$  ratios using a generalized analytical uncertainty equation. The uncertainty equation is given by the Taylor series of the deviations ( $y - y^o$ ) of the output ( $y$ ) from its nominal value ( $y^o$ ) and is expressed in terms of the deviations of the  $i$  inputs ( $x - x_i^o$ ) from their nominal values. As in Morgan and Henrion [1990], for the first three terms, the uncertainty is,

$$\begin{aligned}
 y - y^o &= \sum_{i=1}^N (x_i - x_i^o) \frac{\partial y}{\partial x_i} \bigg|_{X^o} + & \text{I} \\
 \frac{1}{2} \sum_{i=1}^N \sum_{j=1}^N (x_i - x_i^o)(x_j - x_j^o) \frac{\partial^2 y}{\partial x_i \partial x_j} \bigg|_{X^o} + & \text{II} \\
 \frac{1}{3!} \sum_{i=1}^N \sum_{j=1}^N \sum_{k=1}^N (x_i - x_i^o)(x_j - x_j^o)(x_k - x_k^o) \frac{\partial^3 y}{\partial x_i \partial x_j \partial x_k} \bigg|_{X^o} + \dots & \text{III}
 \end{aligned} \tag{5}$$

The subscripts  $X^o$  denote derivatives evaluated at the nominal values. Assuming that there are no covariances between the input variables, all the mixed derivatives in eq. (5) would equal zero. The only terms that would not be zero are the first term and the terms with  $i=j$  and  $i=j=k$  in summations II and III, i.e., second and third derivatives of the variables, respectively. Since the covariances are not known we cannot make the assumption that they are negligible. Given the uncertainties in the variables from which the  $S_a$  ratio is calculated, the uncertainty in  $S_a$  can be estimated without making any assumptions about covariances between inputs. To accomplish this we use Latin

Hypercube Sampling [LHS, *Iman and Conover*, 1980], a statistical sampling method in which a distribution of plausible scenarios of parameter values is generated from a multidimensional distribution. Unlike classical Monte Carlo sampling methods, LHS precludes duplication by requiring that each square grid containing sample positions has only one sample in each row and each column. For this study, we generate 500 variables for each of the seven uncertain parameters used in the calculation of  $S_a$ . We then randomly combine these variables to yield 500 instances or events. Each of these events has a very high probability of yielding a unique  $S_a$  ratio. We then perform standard descriptive statistics on the  $S_a$  values.

The mean and standard deviations of these values is an estimate of the nominal value and uncertainty of dust  $S_a$ . We use the nominal (or central) values in Table 3, suggested by the studies referenced in section 2, to generate the random scenarios. The fine mode radii, coarse mode radii, and imaginary refractive indices are log normally distributed. The fine and coarse geometric standard deviations (GSD), the real refractive indices, and the aspect ratios are normally distributed.

The results obtained by using this method do not assume that the input variables are independent of each other or that  $S_a$  ratio is linear in the individual input variables, i.e., the second- and higher-order derivatives in Equation 5 are not necessarily equal to zero. The statistics of the resulting  $S_a$  ratios provide an uncertainty envelop of the  $S_a$  ratio estimates based on the uncertainty of the inputs. Moreover, the results can be used to explore the sensitivity of the  $S_a$  ratios at each wavelength to the various aerosol

properties. In Figure 24, the 532-nm  $S_a$  ratios are well constrained with a standard deviation of 15% of the mean after perturbing nominal input values shown in Table 3 and similar to the distribution shown in Figure 22. The 1064-nm  $S_a$  ratios are much more sensitive to the addition of noise as shown by the wide spread of 1064-nm  $S_a$  ratios in Figure 24. A parameter that has a significant impact on the  $S_a$  ratios is the complex refractive index. Figure 25 is a 2-D histogram of the 532- and 1064-nm  $S_a$  ratios as functions of the real and imaginary parts of the refractive indices. The range of these values is from 0.00067 to 0.006 for the imaginary part and 1.45 to 1.55 for the real part. The 1064-nm  $S_a$  ratios are sensitive to changes in the refractive index throughout the ranges of these variables. The 532-nm  $S_a$  ratio is sensitive to changes in the complex refractive index at the lower ranges (Figures 25a and 25c). The highest density of 532-nm  $S_a$  ratios are found in midranges of both the real and imaginary part, and these values are insensitive to changes in the complex refractive index. Note that this variation is a total derivative, i.e., all parameters are allowed to vary independently for the scenarios. The 1064-nm  $S_a$  ratios decrease with the complex index of refraction almost monotonically through the range of values (Figures 25b and 25d)



Table 3. Ranges of the variables used to generate 500 random combinations of inputs for T-Matrix calculations. The central values of the size distributions are based on the mean values of the dust layer observed on 08/19/2006 during the NAMMA campaign

Parameter	Minimum	Maximum
Geometric Fine Radius	0.0216	0.194
Geometric Coarse Radius	0.188	1.69
Fine GSD	1.60	1.80
Coarse GSD	1.50	1.70
Real Refractive Index	1.45	1.55
Imaginary Refractive Index	0.00067	0.006
Aspect Ratio	1.70	2.30

## 7. Conclusion

We have determined the  $S_a$  ratios of Saharan dust layers using three methods: transmittance constraint technique for lofted layers, in-situ extinction profile constraint method, and T-Matrix calculations. We found quite robust  $S_a$  ratios at 532 nm and a slightly wider spread at 1064 nm. The three methods yielded 532 nm and 1064 nm  $S_a$  ratios that are quite close.  $S_a$  ratios of  $39.8 \pm 1.4$  sr and  $51.8 \pm 3.6$  sr at 532 nm and 1064 nm, respectively, were produced using the transmittance and 2-color methods applied to CALIPSO measurements of Saharan dust lofted layers. T-Matrix calculations applied to size distributions measured aboard the NASA DC-8 during NAMMA yielded  $S_a$  ratios of  $39.1 \pm 3.5$  sr and  $50.0 \pm 4$  sr at 532 nm and 1064 nm, respectively. The measured extinction profile obtained by the aggregate of nephelometer measurements of total

scattering coefficient and the PSAP measurements of the absorption coefficient was used to constrain the inversion of the CALIPSO measurements. This technique yielded a 532-nm  $S_a$  ratio of 35.7 sr for a dust layer and 25 sr for the aerosol in the marine boundary layer. We perturbed seven microphysical and chemical properties of the dust aerosol and computed the  $S_a$  ratios by randomly combining distributions of these parameters to assess the uncertainty in the  $S_a$  calculation. This uncertainty simulation generated a mean ( $\pm$  uncertainty)  $S_a$  of 39.4 ( $\pm$  5.9) sr and 56.5 ( $\pm$  16.5) sr at 532 nm and 1064 nm, respectively, corresponding to percent uncertainties of 15% and 29%. The ratio of the  $S_a$  ratios,  $S_a(1064 \text{ nm}) / S_a(532 \text{ nm}) = 1.3 \pm 0.13$ , and nearly normally distributed. The simulation revealed that  $S_a$  is insensitive to the middle range of refractive indices at 532 nm and nearly monotonically dependent on the refractive indices at 1064 nm. This explains the observed robust  $S_a$  ratios at 532 nm and relatively wide spread of  $S_a$  at 1064 nm.

This study examined a wide range of dust loadings in various locations within ~1250 km of Saharan Desert source regions on different days. The  $S_a$  ratios do not change significantly with dust loading, suggesting the dust microphysical and chemical properties do not vary appreciably. The profile of  $S_a$  ratios at 532 nm are very close to 40 sr using different methods and on different days. There is a wider variation in the 1064 nm  $S_a$  ratios for all methods. It is possible that the variation of  $S_a$  at 1064 nm is driven by a higher sensitivity to changes in the complex index of refraction at the 1064 nm wavelength. The ratio of  $S_a$  ratios at the two wavelengths [ $S_a(1064 \text{ nm})/S_a(532 \text{ nm})$ ] is  $1.3 \pm 0.13$ .

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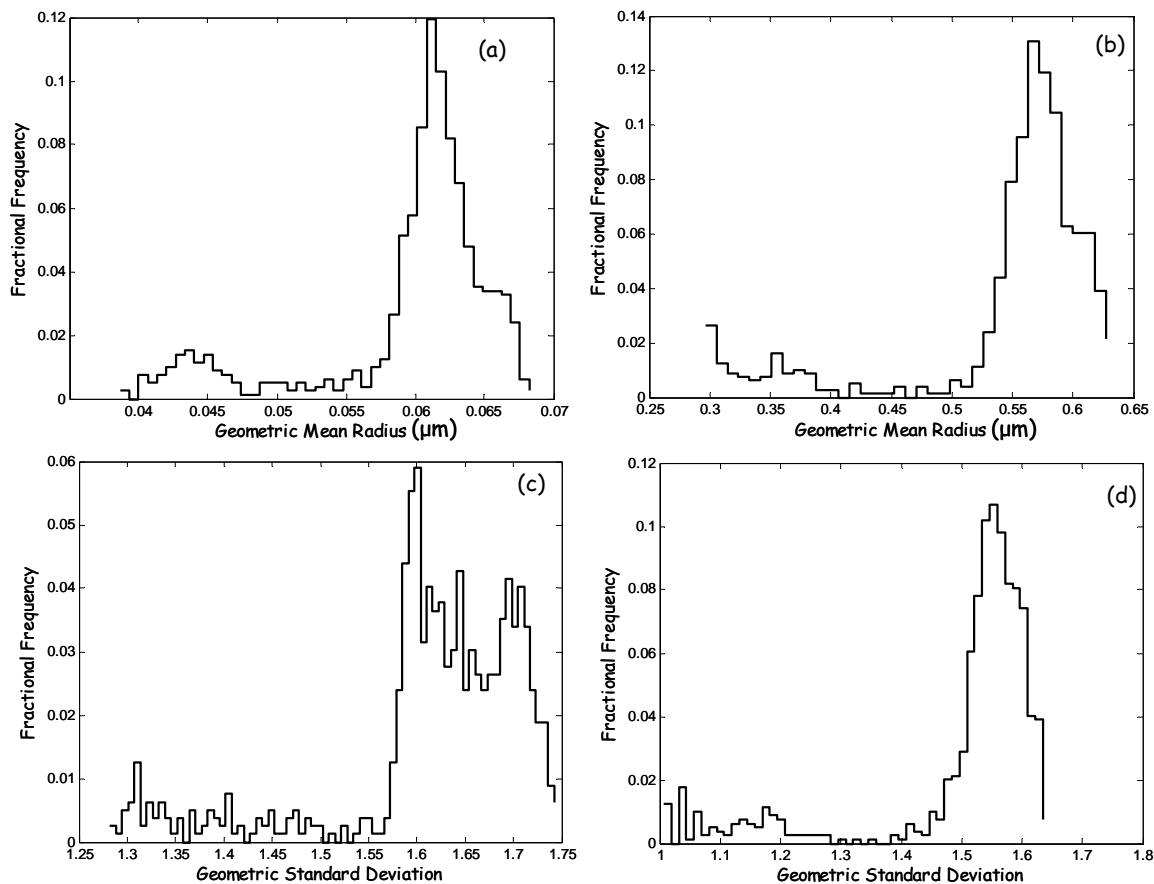
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Figure 1. (a) A Two-dimensional probability plot of NAMMA in situ size distributions measured in a dust layer over a 22 minute period (~260 size distributions) during Flight 4 on August 19, 2006. The mean measured size distribution is denoted by the yellow line, and (b) the best approximate bimodal log-normal particle size distribution (blue squares) with fine (coarse) mean radius of 0.0648 (0.5627)  $\mu\text{m}$  and a fine (coarse) geometric standard deviation of 1.696 (1.572) fitted to the average measured size distribution (red squares).

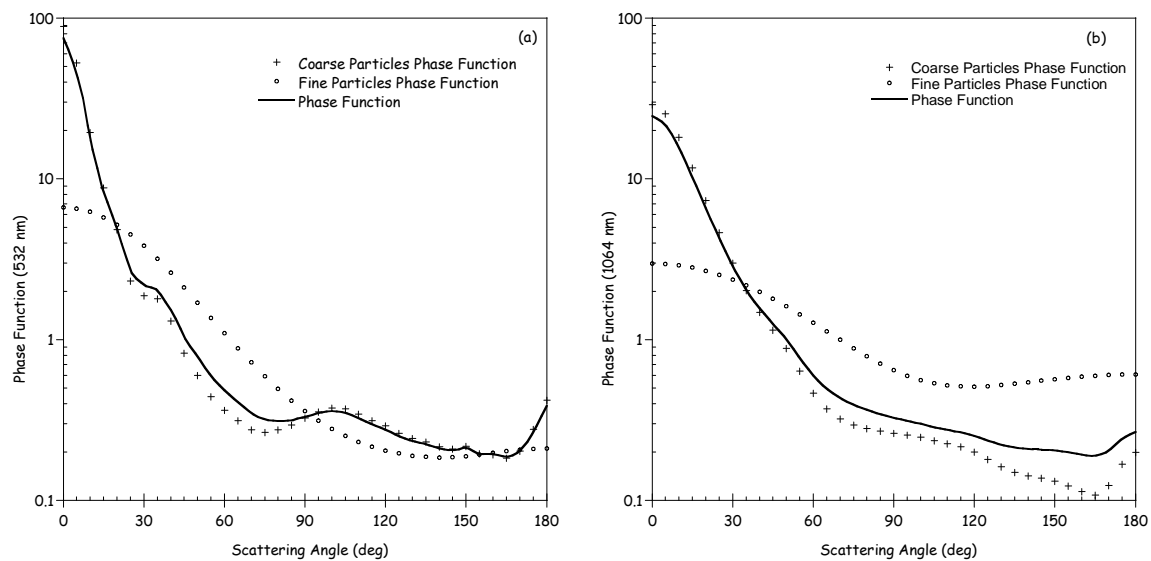


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997 Figure 2. The distributions of the fine (a) and coarse (b) geometric mean radii and the  
 998 fine (c) and coarse (d) geometric standard deviations for a dense elevated dust layer  
 999 measured during a flight on August 25, 2006.

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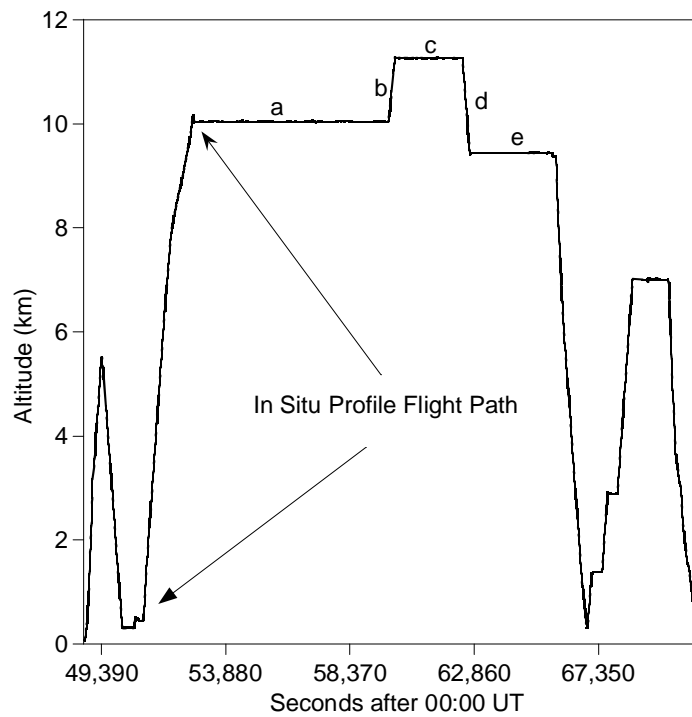
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1003 Figure 3. The (a) 532-nm and (b) 1064 nm Saharan dust phase functions calculated from  
 1004 the size distributions measured aboard NASA DC-8 during Flight 4 on August 19, 2006.  
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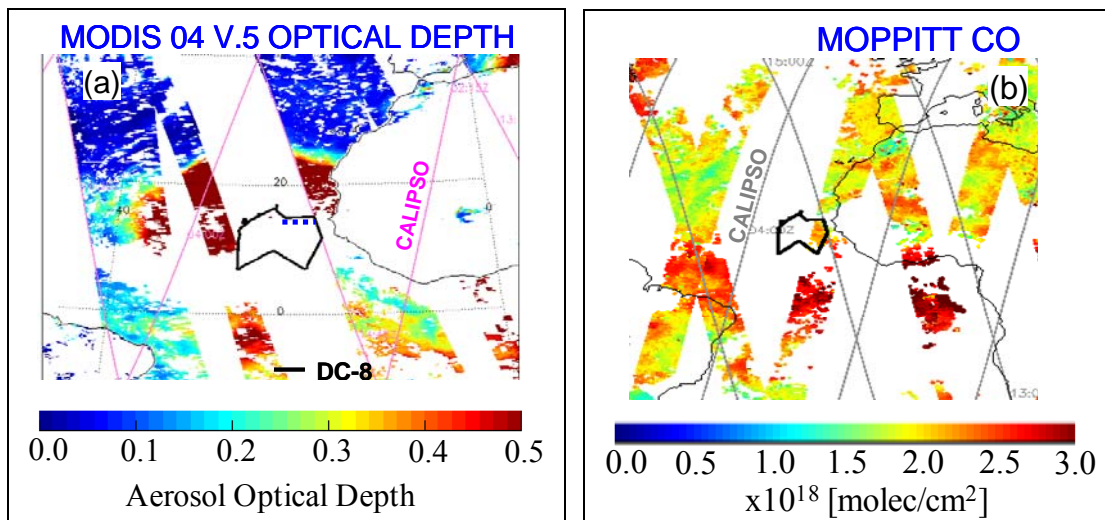
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1008 Figure 4. The DC-8 altitudinal flight path showing the segment during which the in situ  
1009 measurements were made. The direct CALIPSO underflights are the five segments (a, b,  
1010 c, d, and e) at the highest altitudes. The time is in seconds after midnight UTC.  
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1014 Figure 5. CALIPSO flight tracks superimposed on (a) the MODIS optical depth and (b)  
 1015 the MOPPITT CO concentrations. The DC-8 flight track for August 19, 2006 is shown  
 1016 by the black lines in the middle of both figures. The blue dots in (a) denote the portion of  
 1017 the DC-8 flight segment during which the in-situ measurements were taken. Daytime  
 1018 CALIPSO tracks run from the southeast to the northwest while the nighttime tracks run  
 1019 from northeast to the southwest.  
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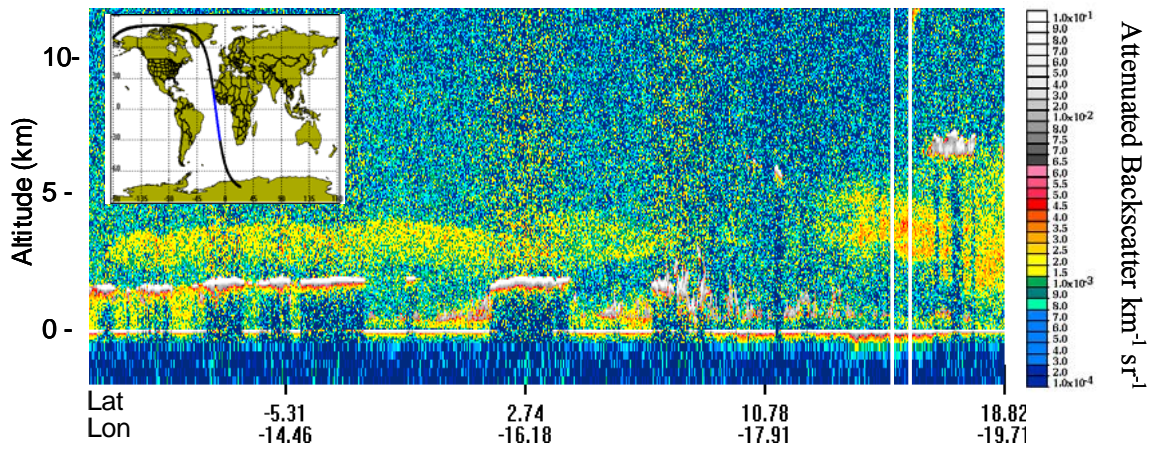
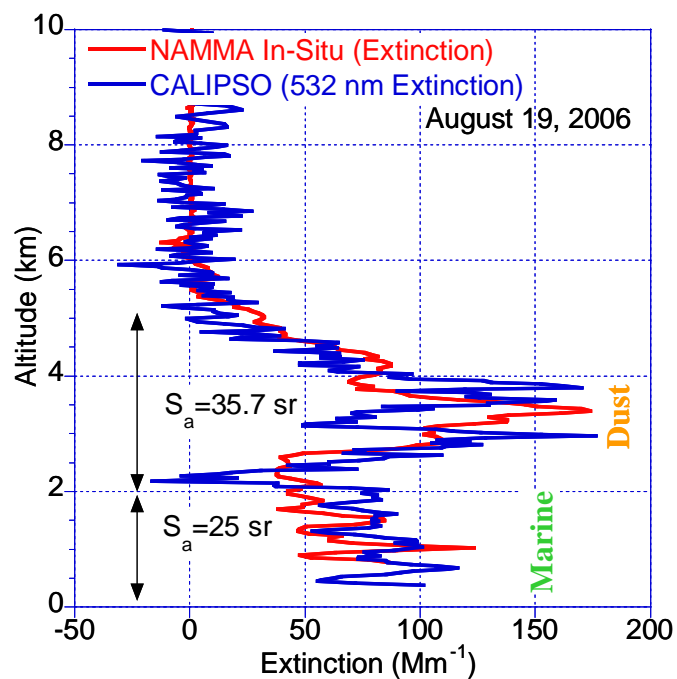


Figure 6. Browse image of the CALIPSO flight track on August 19, 2006 shown in blue in the inset map and the location of the near coincidence of the CALIPSO flight track and the DC-8 flight 4 during NAMMA is denoted by the two white lines near 15N, 19W. In the image, colors above the surface are coded as follows: blue indicates clear sky, white indicates clouds, and warm colors (green, yellow, orange, and red) indicate aerosols. The white parallel lines bound the CALIPSO profiles closest to the in-situ measurements.

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1032 Figure 7. A comparison of the profiles of the in situ extinction coefficient derived during  
 1033 the DC-8 ascending segment as shown in Figure 4 and the extinction coefficient retrieved  
 1034 from the CALIPSO measurement averaged over the region indicated by the two white  
 1035 lines in Figure 6. The  $S_a$  ratios were determined using the in situ extinction as an  
 1036 additional constraint.  
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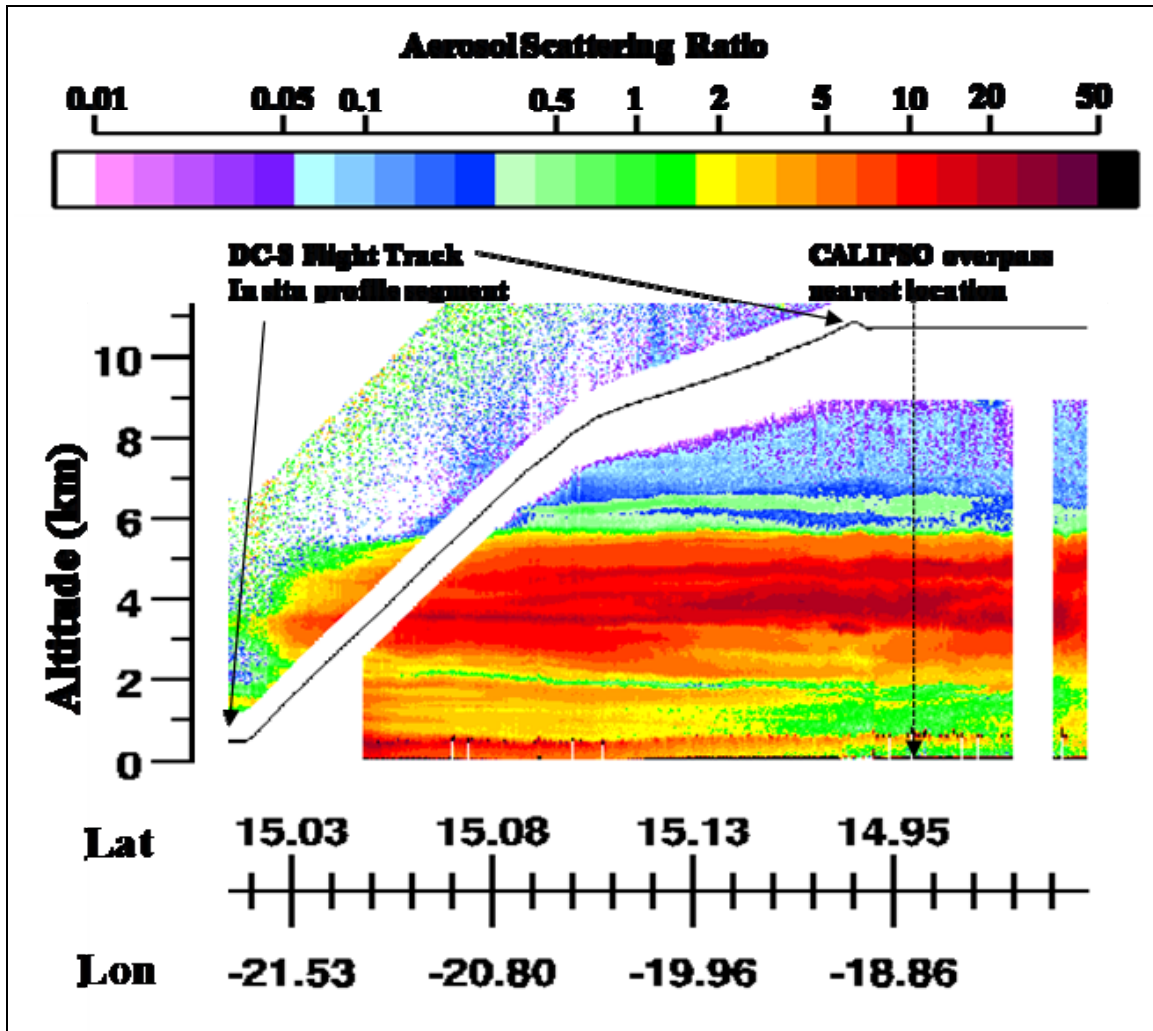


Figure 8. Aerosol scattering ratio ( $\beta_a/\beta_m$  at 815 nm) image measured by the Lidar Atmospheric Sensing Experiment (LASE) aboard DC-8 during the ascending segment passing through a Sahara Air Layer (SAL) and the underflight of CALIPSO as shown in Figure 4. The DC-8 point of nearest approach to the CALIPSO flight track is near 15°N-19°W.

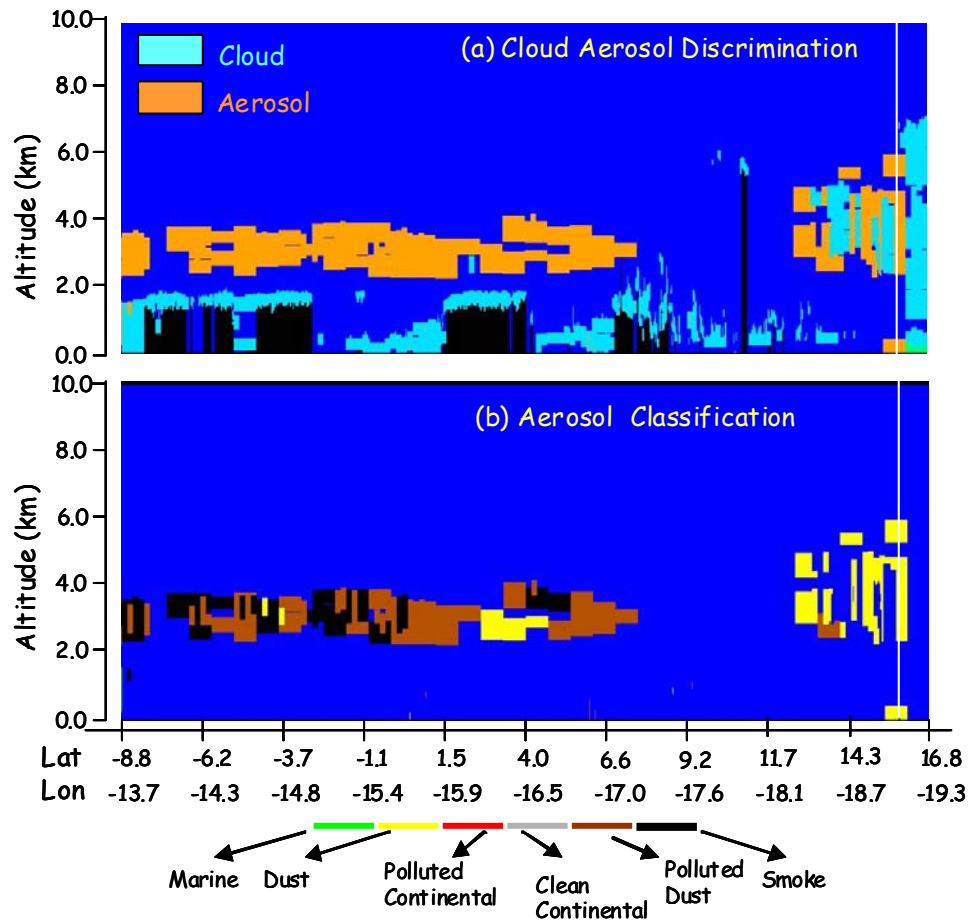
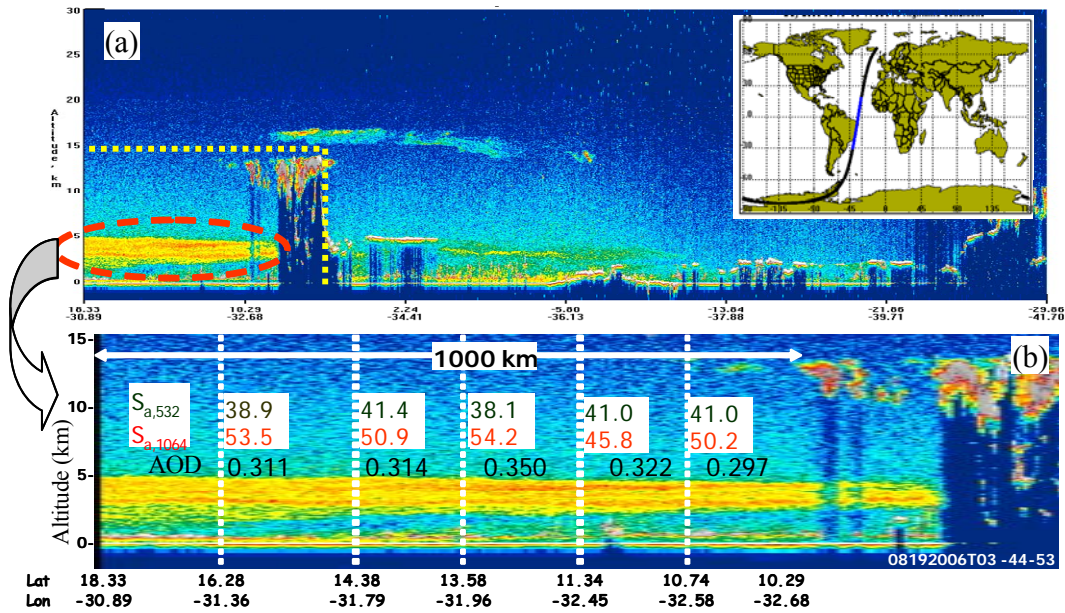
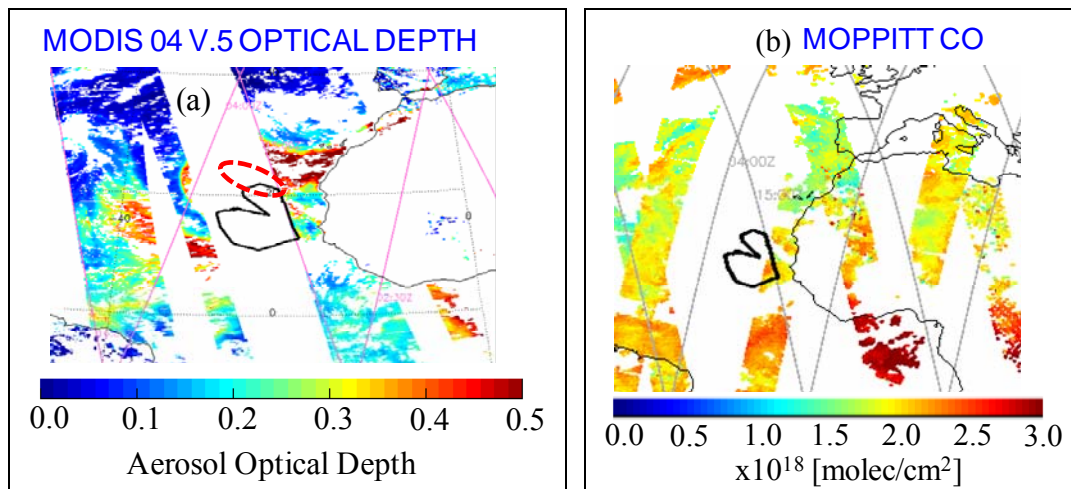


Figure 9. Products of the CALIPSO (a) cloud-aerosol discrimination and (b) aerosol classification schemes for the data in the browse image shown in Figure 6. The cloud aerosol discrimination shows the extensive aerosol layer at 3 km and the aerosol classification image shows that this aerosol is a Saharan dust layer to the north and a polluted dust and smoke layer to the south.



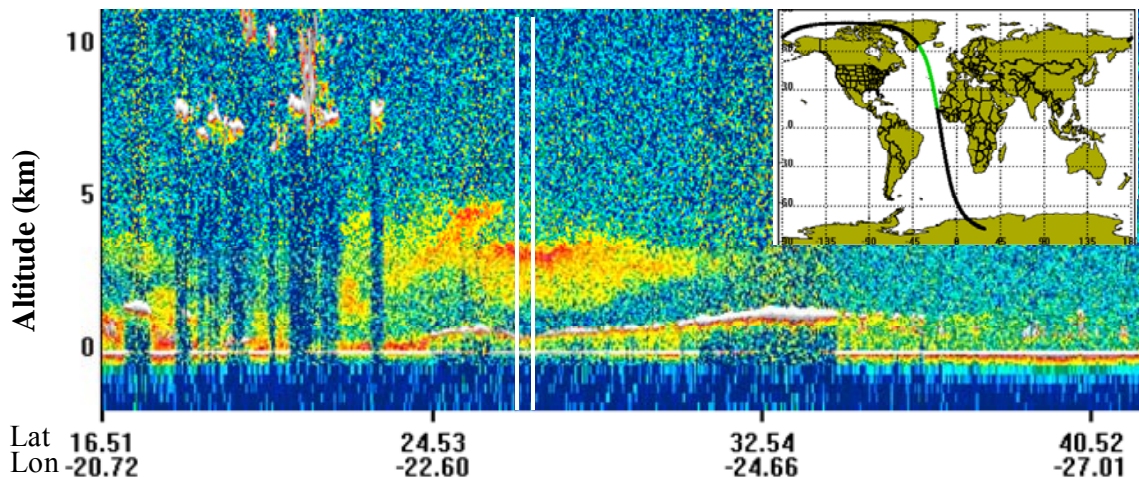
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1061 Figure 11. CALIPSO flight tracks superimposed on the (a) MODIS optical depth and the  
 1062 (b) MOPPITT CO concentrations. The CALIPSO flight tracks are colored cyan and grey  
 1063 in the MODIS and MOPPITT images, respectively. The DC-8 flight track for August 26,  
 1064 2006 is shown by the heart shaped drawing in the middle of both figures. The CALIPSO  
 1065 underflight segment is the portion of the flight track in alignment with the CALIPSO  
 1066 flight track in cyan or grey in (a) or (b), respectively. The DC-8 in situ data for this  
 1067 obtained during the descent flight segment shown by a red dashed tilted oval in (a).  
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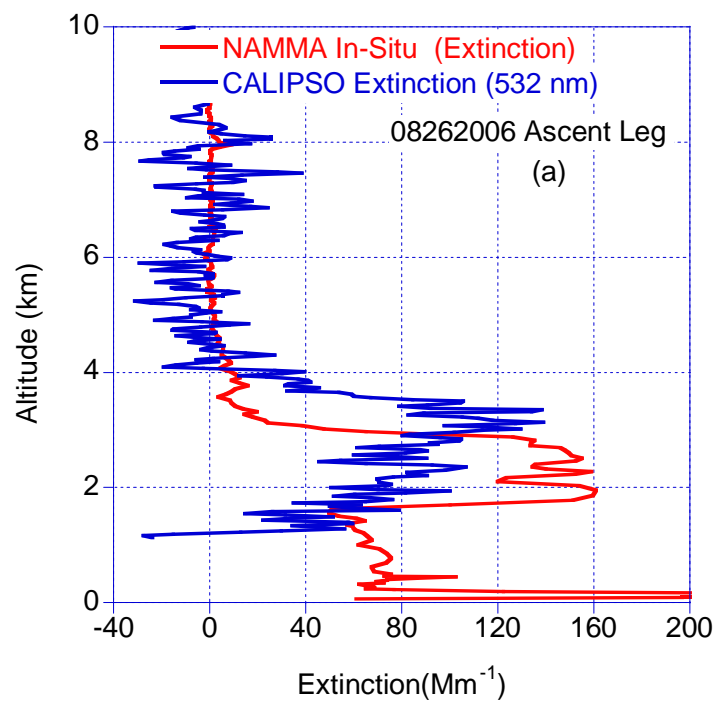
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1071 Figure 12. Browse image of the CALIPSO flight track shown in green in the inset map  
 1072 and the location of the near coincidence (in space) of the CALIPSO flight track and the  
 1073 DC-8 flight 8 on August 26, 2006. The color code is as in Fig. 6. The white lines denote  
 1074 the CALIPSO data used for comparison with the DC-8 measurements.  
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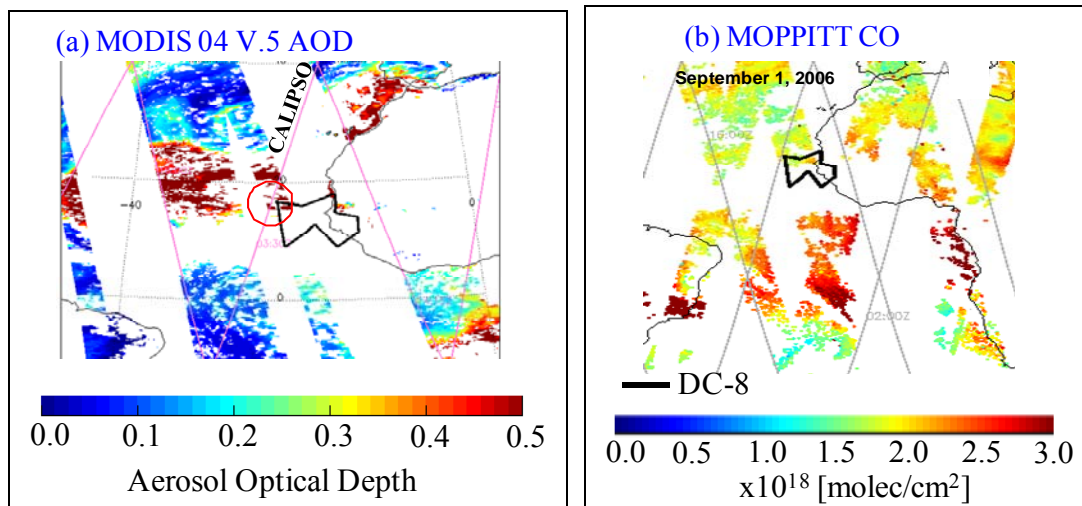


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1078 Figure 13. A comparison of the NAMMA in situ extinction profile and the extinction  
1079 profile calculated from CALIPSO's attenuated backscatter measurement using 532 nm  $S_a$   
1080 ratio of 38.2 sr obtained using the transmittance method. The two measurements both  
1081 taken on August 26, 2006 are offset by 2 hours and ~1250 km.  
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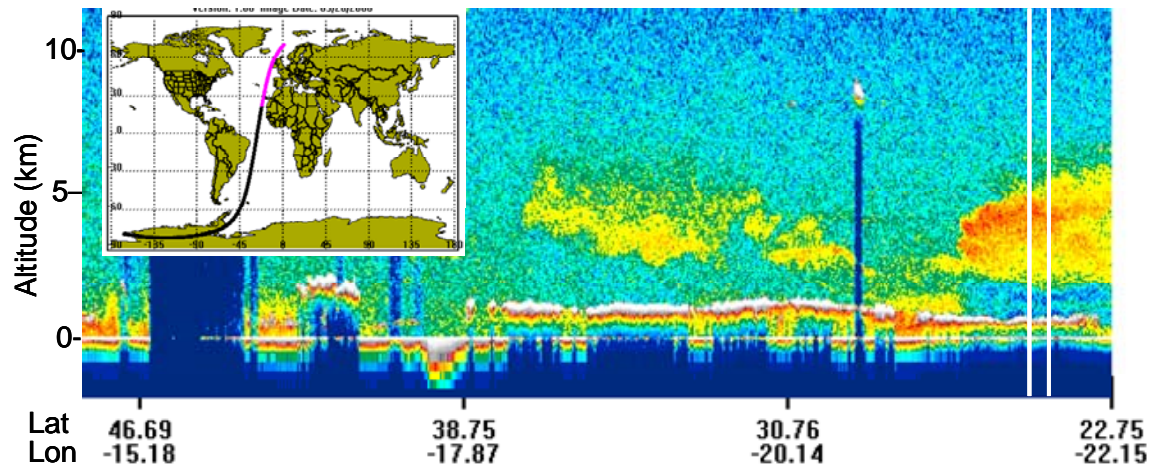
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1085 Figure 14. CALIPSO flight tracks superimposed on the (a) MODIS optical depth and the  
1086 (b) MOPPITT CO concentrations. The CALIPSO flight tracks are colored cyan and grey  
1087 in the MODIS and MOPPITT images. The DC-8 flight track for September 1, 2006 is  
1088 shown by the black lines in the middle of both figures.  
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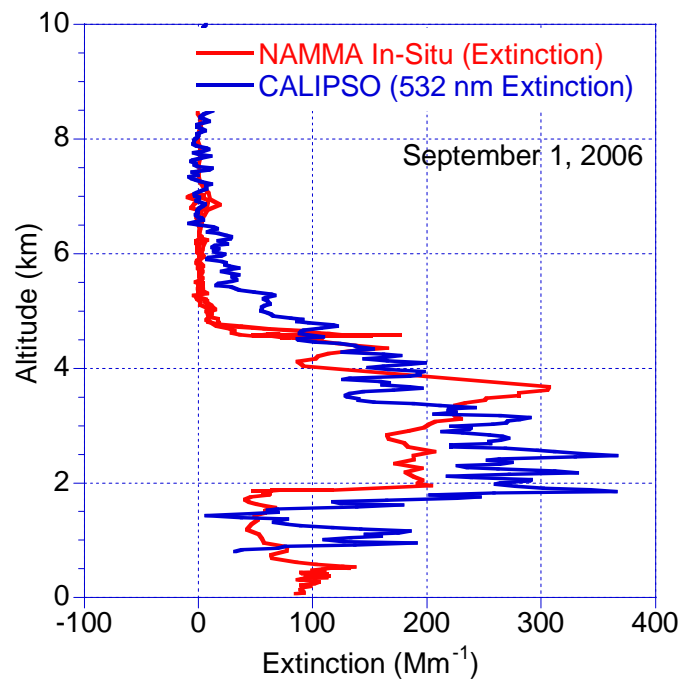


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1092 Figure 15. Browse image of the CALIPSO flight track shown in cyan in the inset map  
1093 and the location of the near coincidence (in space) of the CALIPSO and the DC-8 flight  
1094 tracks on September 1 during NAMMA. The color code is as in Fig. 6. The white lines  
1095 subtend the region averaged for the extinction comparison.  
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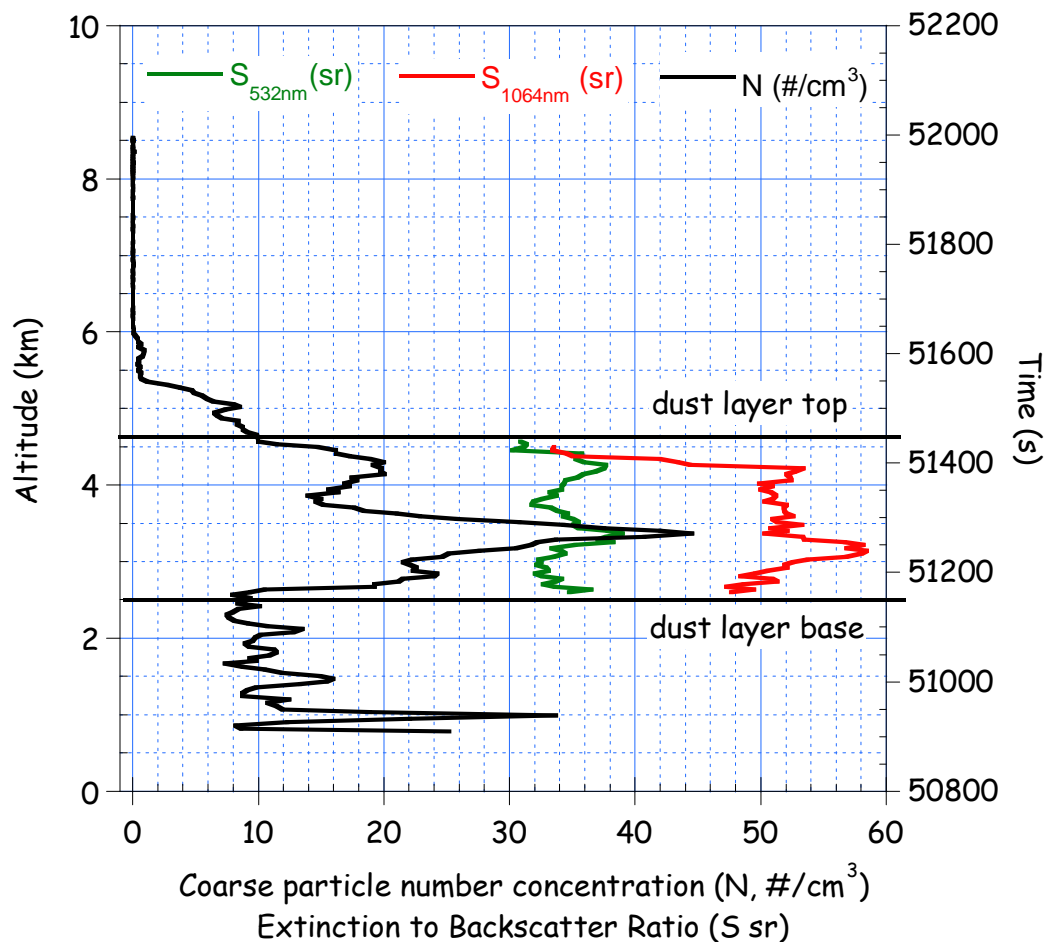


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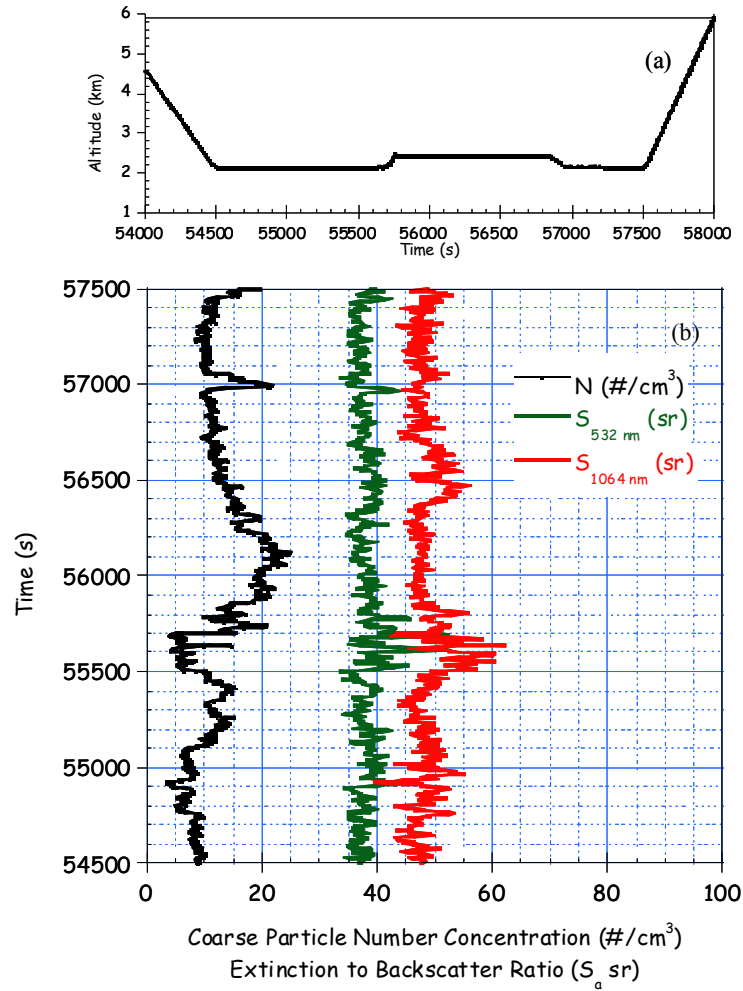
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1099 Figure 16. The NAMMA/CALIPSO extinction comparison for Flight 10 on September 1,  
1100 2006. Note that the centroid of the layer has shifted upwards by about 1.5 km in between  
1101 the CALIPSO measurement and NAMMA sampling. The CALIPSO measurement is ~15  
1102 hours earlier and nearly 750 km away.  
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1104

1105 Figure 17. A profile of the 1064 and 532-nm  $S_a$  ratios and integrated number distribution  
 1106 of coarse particles (0.583  $\mu\text{m}$  – 6.264  $\mu\text{m}$ ) taken from a 20-minute segment of the APS  
 1107 measurements during Flight 4 on August 19, 2006. The dust base and top are shown by  
 1108 the lines at altitudes of 2.5 km and 4.6 km, respectively. The mean 532-nm and 1064-nm  
 1109  $S_a$  ratios for the layer is  $34.3 \pm 2.0$  sr and  $50.2 \pm 5.7$  sr, respectively.  
 1110



1111

1112 Figure 18. (a) The flight track of the DC-8 on August 25, 2006. The straight and nearly  
 1113 level portion is through a dust layer at ~2 km. (b) The calculated 532- and 1064-nm  $S_a$   
 1114 ratios and coarse number particle concentration for the straight and level portion of this  
 1115 flight.  
 1116

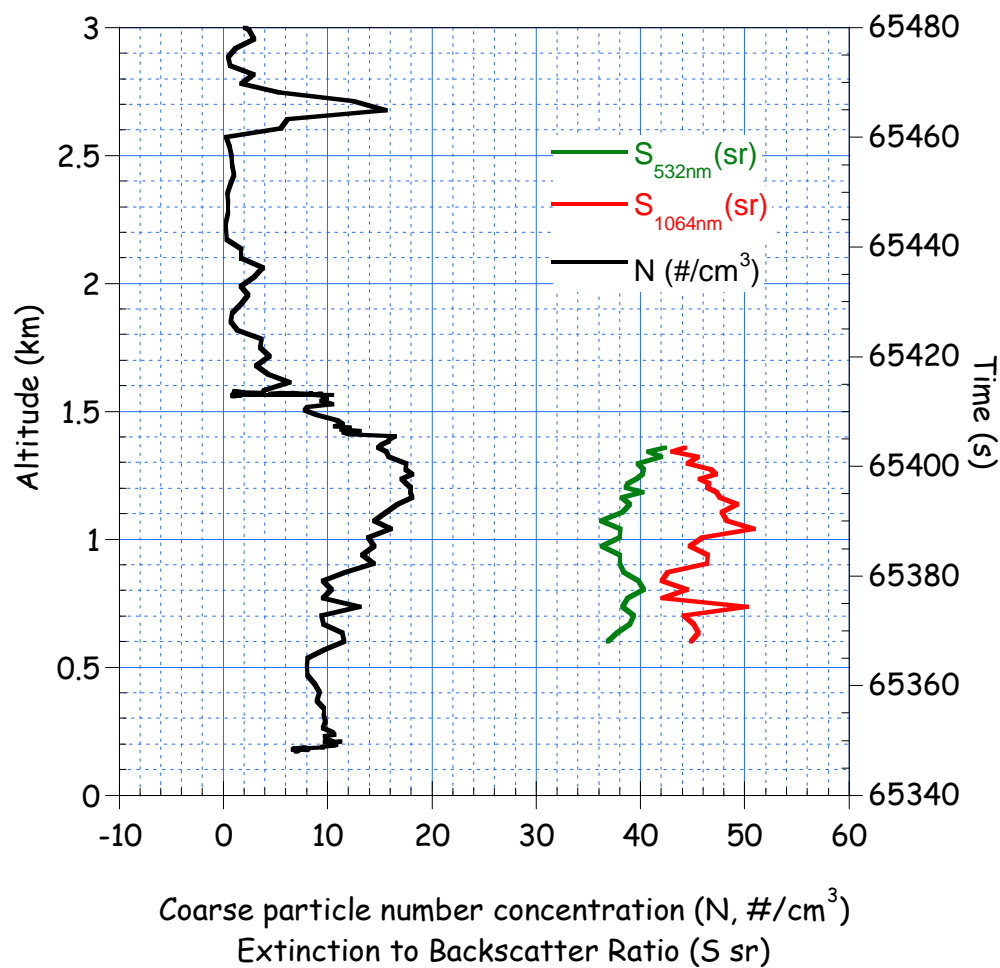
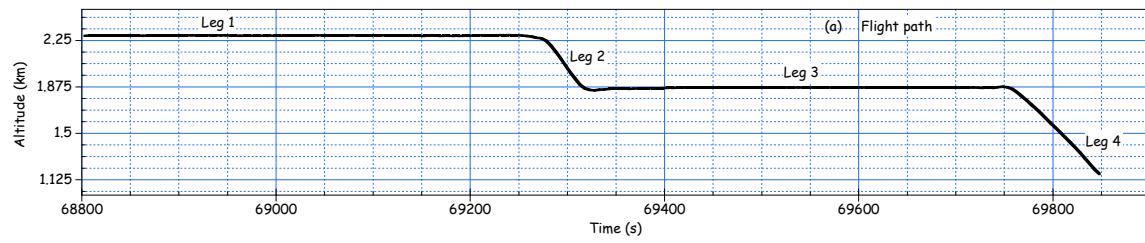
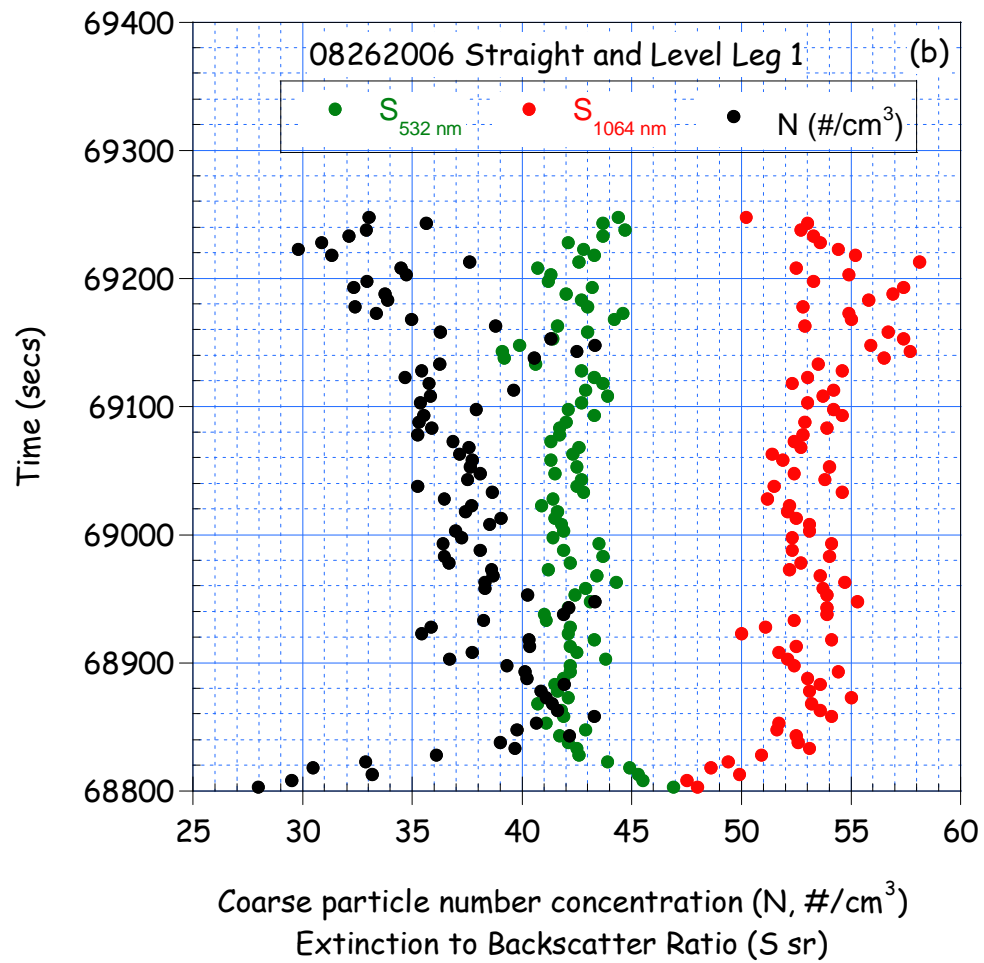


Figure 19. A profile of the 1064- and 532 -nm  $S_a$  ratios, and integrated number distribution of coarse particles during Flight 8 on Aug. 26, 2006. The mean 532-nm and 1064-nm  $S_a$  ratios for the layer is  $39.0 \pm 1.5$  sr and  $45.9 \pm 2.2$  sr, respectively.

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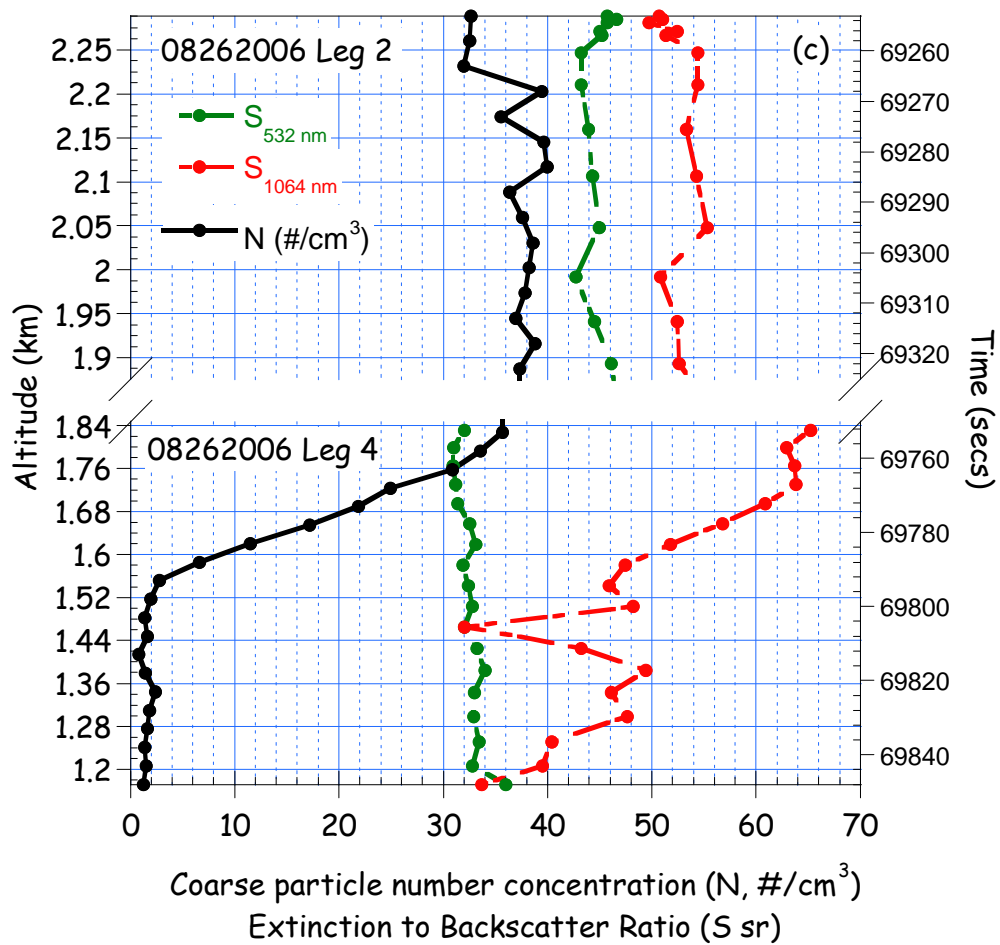


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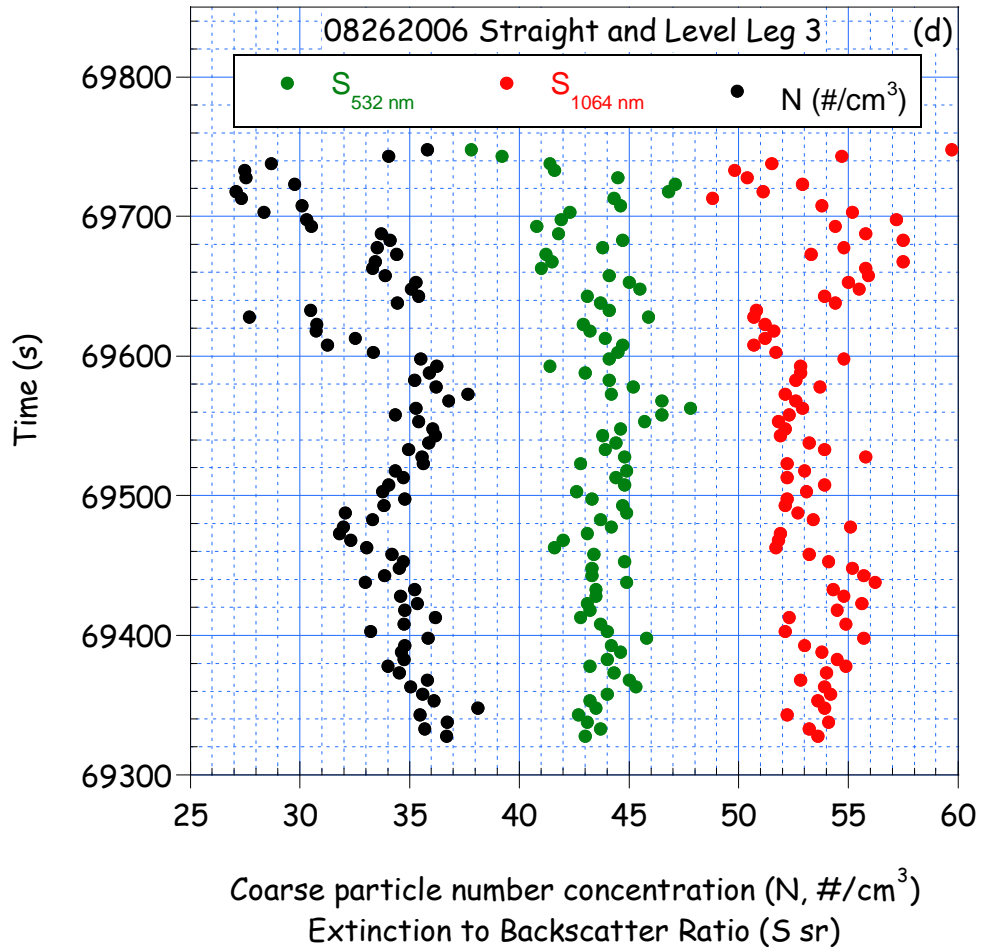
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1129 Figure 20 . (a) Flight path and  $S_a$  ratios and the particle number concentration for the  
 1130 return flight on August 26, 2006. The figures are plotted by temporal profiles for the  
 1131 straight and level legs, and vertical profiles for descent legs. (b) is temporal profile of  
 1132 straight and level leg 1, (c) vertical profiles of descent legs 2 and 4, and (d) temporal  
 1133 profile of straight and level leg 3.  
 1134

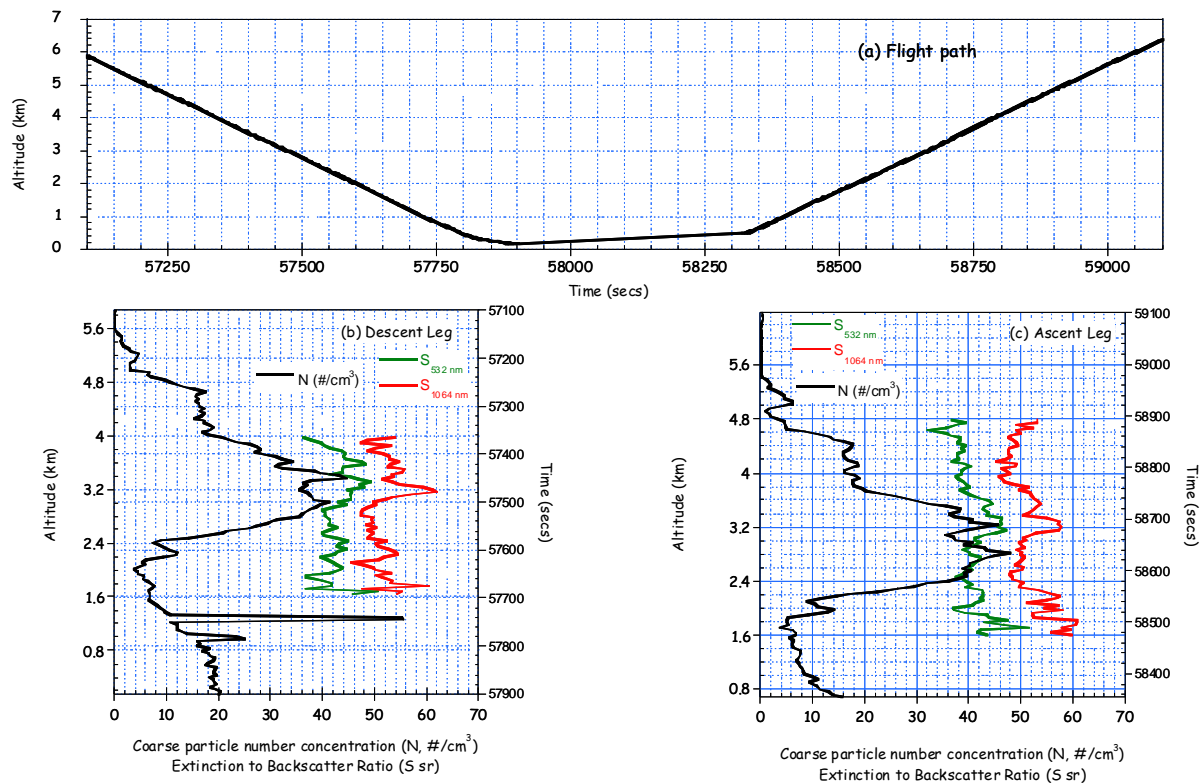
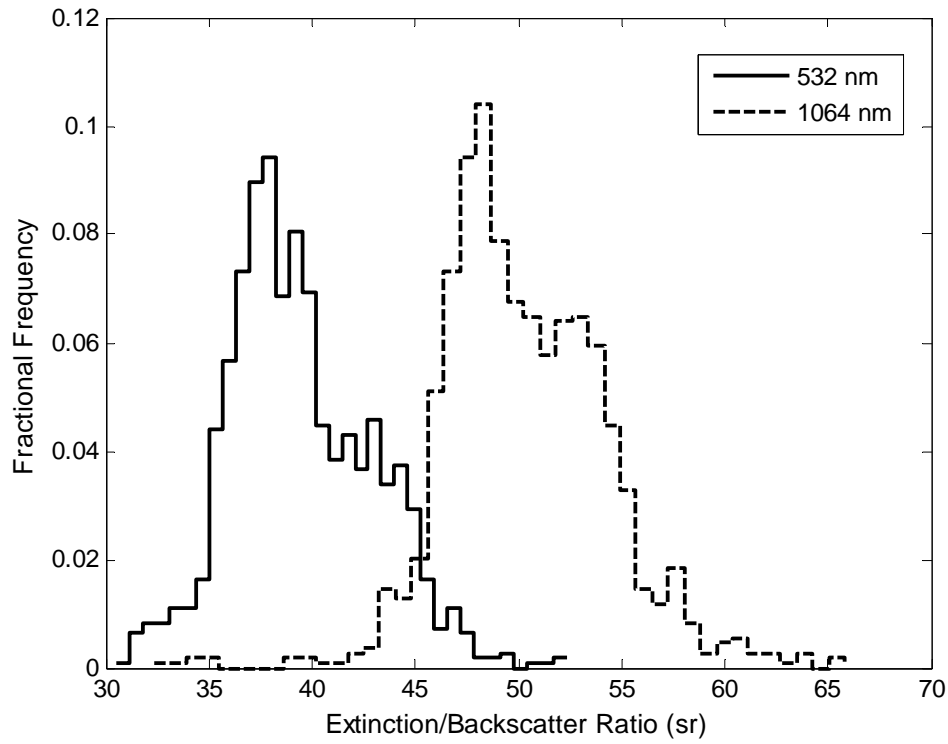


Figure 21. The (a) flight path and  $S_a$  ratios and the particle number concentration for the flight on August 20, 2006 plotted vertical profiles of (b) the descent leg and (c) the ascent leg.

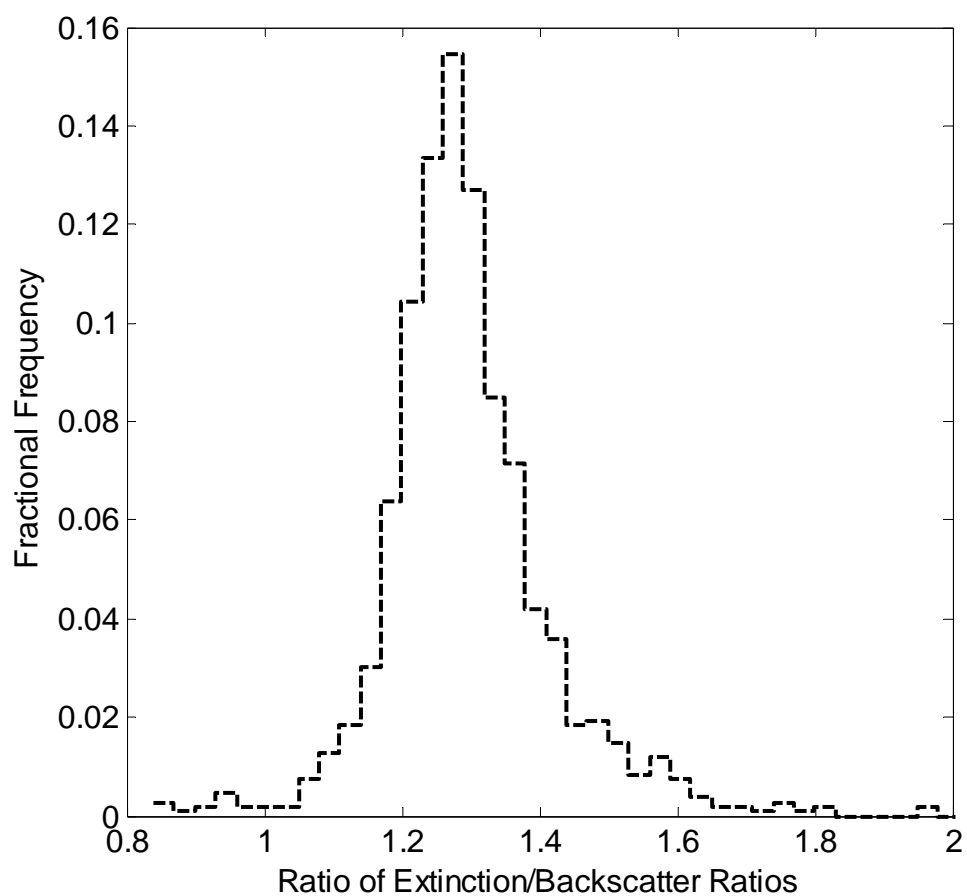




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1141 Figure 22. An ensemble of all the  $S_a$  ratio estimates (~1100 records) based on the  
 1142 NAMMA in-situ size distributions of dust layers and T-Matrix calculations. The average  
 1143  $S_a$  ratios are  $39.1 \pm 3.5$  sr and  $50.0 \pm 4$  sr at 532 nm and 1064 nm, respectively  
 1144

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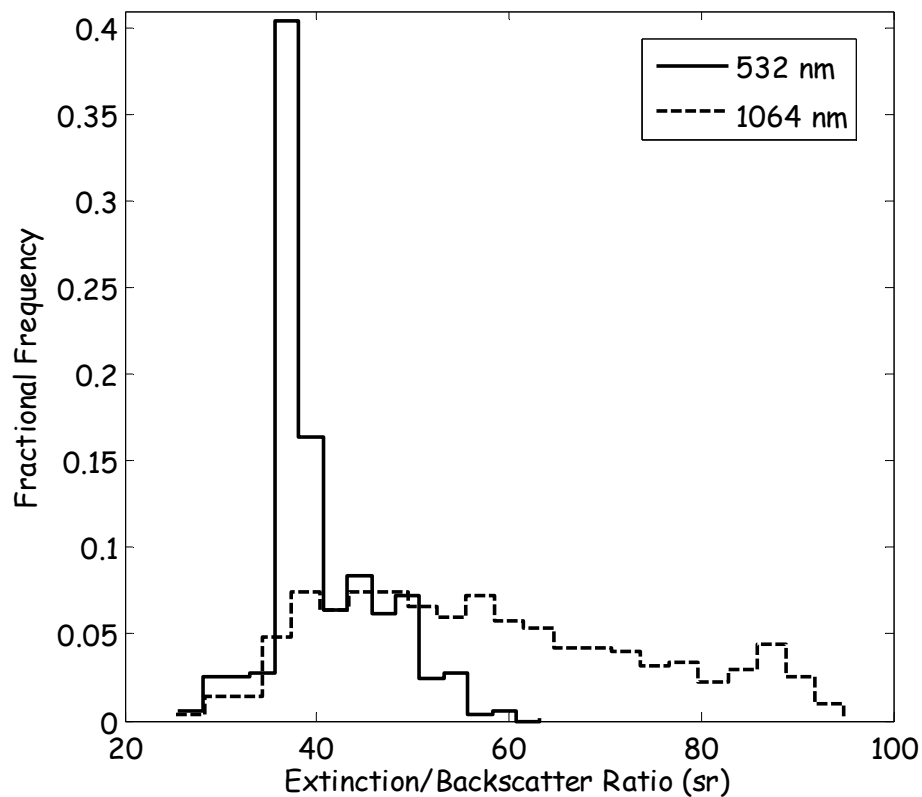
1147 Figure 23. The ratio of the  $S_a$  ratios [ $S_a$  (1064 nm)/ $S_a$  (532 nm)] derived from the size  
1148 distribution data used for this study.

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1155 Figure 24. The distribution of the  $S_a$  ratio calculated from a Latin Hypercube Sample  
 1156 (LHS) of 500 events bounded by the ranges shown in Table 3. The normal means ( $\pm 1$   
 1157 standard deviation) of the 532- and 1064-nm  $S_a$  ratios are  $39.4 \pm 5.9$  sr and  $56.5 \pm 16.5$  sr,  
 1158 respectively.

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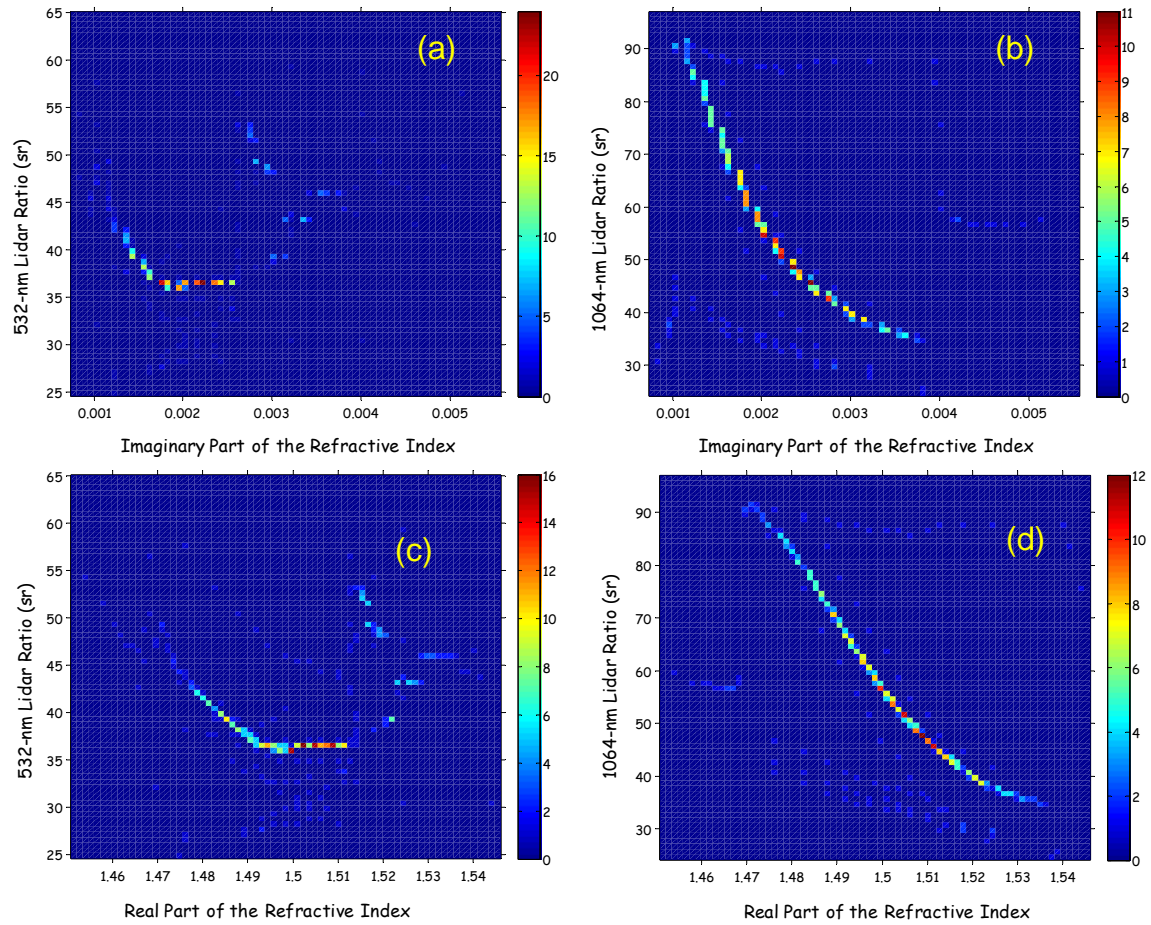


Figure 25. Two-dimensional frequency plots showing the variation of the lidar ratios with real and imaginary parts of the complex refractive indices for 500 randomly generated events discussed in section 6.