Aerosol Size Distribution, Particle Concentration, and Optical Property Variability near Caribbean Trade Cumulus Clouds: Isolating Effects of Vertical Transport and Cloud Processing from Humidification Using Aircraft Measurements

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ABSTRACT

This paper examines the effect of trade wind cumulus clouds on aerosol properties in the near-cloud environment using data from the Rain in Cumulus over the Ocean (RICO) campaign. Aerosol size distributions, particle concentrations, and optical properties are examined as a function of altitude and distance from cloud, at ambient relative humidity (RH) and adjusted to a constant RH to isolate effects of humidification from other processes.

The cloud humidity halo extended about 1500–2000 m from the cloud edge, with no clear altitude dependence on horizontal extent over an altitude range of 600–1700 m. The combined effects of vertical transport of aerosol by clouds and cloud processing contributed to the modification of aerosol size distributions within the clouds' humidity halos, particularly close to the cloud boundaries. Backscatter at 532 nm, calculated from the aerosol properties, exhibited no distinguishable trend with altitude within 400 m of cloud edges, increased toward lower altitudes beyond 400 m, and decreased away from cloud boundaries at all altitudes. The mean aerosol diameter was found to rapidly decline from 0.8 to 0.4 μm from near the cloud boundary to the boundary of the humidity halo. Aerosol optical depth at 532 nm within the layer between 600 and 1700 m increased near exponentially from 0.02 to 0.2 toward the cloud boundaries within the humidity halo. These trends agreed qualitatively with past space-based lidar measurements of trade wind cloud margins, although quantitative differences were noted that likely arose from different sampling strategies and other factors.

1. Introduction

Space-based estimation of the direct shortwave radiative aerosol effect and forcing on the climate system (e.g., Loeb and Kato 2002; Sekiguchi et al. 2003; Loeb and Manalo-Smith 2005; Remer and Kaufman 2006) depends on accurate retrieval of aerosol optical properties from remote sensors. Determination of aerosol optical properties is normally limited to clear-air regions, with cloud-masking algorithms (e.g., Higurashi and Nakajima 1999; Martins et al. 2002; Hutchison et al. 2008) used to distinguish clear from cloudy pixels in satellite images. The notion that clear and cloudy regions cannot easily be separated has been raised in past studies (e.g., Wielicki and Welch 1986; Yang and Di Girolamo 2008) and has recently come under more scrutiny because of effects of optically thin cloud fragments, humidification, and growth of aerosols in the near-cloud environment and 3D cloud-radiative transfer effects (Wen et al. 2007; Charlson et al. 2007; Koren et al. 2007; Yang and Di Girolamo 2008; Marshak et al. 2008). The problem is particularly severe in regions of the atmosphere with heavy aerosol loading but can even extend to finer scales associated with individual cumulus clouds (e.g., Tackett and Di Girolamo 2009, (hereafter TD09); Twohy et al. 2009).

Trade wind cumuli are ubiquitous throughout the marine boundary layer across much of the world’s tropical oceanic regions. Statistical analyses of the distribution of trade wind cumuli, based on Advanced Spaceborne Thermal Emission and Reflection Radiometer (ASTER) 15-m-resolution imagery over the Atlantic east of the Caribbean Sea, shows that 75% of these clouds have a nearest neighbor within 10 times their area-equivalent radius, which is equivalent to clear points being within...
5000 m of cloud edges 90% of the time, given the observed distributions (Zhao and Di Girolamo 2007). Cloud Aerosol Lidar with Orthogonal Polarization (CALIOP) measurements over the Indian Ocean indicate that 66% of cloud free regions are within 4000 m of a nearby cloud (Twory et al. 2009). It is well documented that cumulus clouds frequently exhibit enhanced humidity within one to two cloud radii beyond the cloud edge depending on shear, cloud age, and the presence of stable layers (Radke and Hobbs 1991; Perry and Hobbs 1996; Lu et al. 2003; Laird 2005; Twohy et al. 2009). The humidity enhancement develops as turbulence at the cloud boundaries, because of evaporation and subsidence, laterally mixes cloudy air with the unsaturated air in the near cloud environment (Rodts et al. 2003; Heus and Jonker 2008; Heus et al. 2009; Wang et al. 2009; Wang and Geerts 2010). The existence of higher humidity near clouds, together with the cloud-proximity findings of Zhao and Di Girolamo (2007) and Twohy et al. (2009), imply that the aerosol characteristics within the cloud margin—the zone encompassing a few kilometers beyond the cloud boundaries—may be needed to accurately determine the direct shortwave radiative aerosol effect in clear air over much of the tropical oceans.

It is well known that precipitation within clouds selectively removes atmospheric aerosol altering the residual aerosol size distribution. Clouds transport aerosol vertically (e.g., Rodhe 1983; Kleinman and Daum 1991; Cautenet and Lefeuvre 1994; Yin et al. 2005) and chemical processes within cloud drops (which we refer to herein as cloud processing) can alter the size and composition of aerosol particles (e.g., Easter and Hobbs 1974; Hegg and Hobbs 1982; Flossmann et al. 1987; Hoppel et al. 1990, 1994; Leaitch 1996; Bower et al. 1997). Aerosol processed by clouds are deposited into the cloud margin as entrainment and lateral mixing lead to evaporation of cloud droplets and transport of the residue outward into the cloud margin. Together, cloud processing, vertical transport, detrainment of moisture and aerosol, and turbulent mixing might be expected to lead to differences in concentration, size, composition, and optical properties of aerosol within cloud margins compared to clear air far from clouds (e.g., Hoppel et al. 1986; Radke and Hobbs 1991; Perry and Hobbs 1994; Flossmann 1998; Lu et al. 2003; Peter et al. 2006; Roelofs and Kamphuis 2009; Twohy et al. 2009). Indeed, remote sensing studies of aerosol optical properties within cloud margins using a variety of instrumentation platforms [e.g., surface and aircraft-based sun photometers (Koren et al. 2007; Redemann et al. 2009), the space-based CALIOP (TD09) and an aircraft-mounted Multichannel Cloud Radiometer (Twohy et al. 2009)] all have found greater aerosol extinction within cloud margins. Based on measurements of aerosol made in trade wind cumuli over the Indian Ocean, Twohy et al. (2009) argue that increased humidity (and associated larger equilibrium sizes of haze droplets), rather than changes in aerosol concentration, are primarily responsible for the enhancement in aerosol extinction. Their conclusion was based on evidence that particle concentrations, measured over the size range 0.01–20 μm, were largely invariant as a function of distance from the clouds, except in the first 100 m where potential cloud contamination rendered larger uncertainties in their data. However, TD09 argued that humidity variations alone are not enough to explain the larger aerosol backscatter and color ratio observed by CALIOP, suggesting that additional processes must also be included.

The work presented herein further examines the effect of clouds on aerosol particle distributions in their vicinity. This study uses data from a large number of trade wind cloud penetrations made during the Rain in Cumulus over the Ocean (RICO) experiment (Rauber et al. 2007). This work is unique in that particle size distributions and concentrations are examined in the cloud environment as a function of altitude above cloud base and distance from cloud, adjusted to a constant relative humidity (RH) to isolate the effects of vertical transport and cloud processing from humidification. The relative humidity distribution is also analyzed to compare the current data with past work. Finally, observed particle size distributions at ambient RH are used to examine optical properties of aerosol as a function of distance from cloud and altitude within the cloud layer. Results are compared with recent satellite-based CALIOP observations reported by TD09 over the same tropical region.

2. Data sources and methodology

The data used in this study were obtained during the RICO field campaign. The study area was over the Atlantic Ocean just northeast of the Caribbean island of Barbuda during the winter months of 2004/05 (Rauber et al. 2007). Thermodynamic, moisture, environmental, and rainfall characteristics of the RICO region during the field campaign can be found in Davison et al. (2013a,b,c), Snodgrass et al. (2009), Nuijens et al. (2009), and Minor et al. (2011). There were 19 research flights carried out with the National Center for Atmospheric Research (NCAR) C-130 (Rauber et al. 2007); 5 flights (RF01, RF02, RF06, RF07, and RF19) were eliminated from this study either because of instrumentation problems or because the flight was not focused on sampling trade wind cumuli. All flights used in this study were approximately 8 h long. The first and last 2 h of each flight were dedicated to sampling along three 60-km-diameter
circles flown at 100 m (just above the ocean), about 400 m (just below cloud base), and 4600 m (above clouds to release dropsondes). The central 4 h of each flight were used to sample trade wind cumuli. The data included in this study were from the central 4 h of the 14 included flights and were limited to cloud penetrations between 600 and 1700 m above the ocean surface. Although clouds were occasionally sampled at higher altitudes, the amount of data was insufficient to characterize the aerosol size distributions. In addition, air sampled during the higher tracks was often a mixture of air from above and within the trade wind layer, leading to complications in sorting the effect of cloud detrainment from mixing across air masses. The exact start and end time (UTC) of the data used for each flight are given in Table 1.

RICO focused on statistical properties of trade wind cumuli, rather than process studies. Individual clouds were rarely sampled more than once. Here we adopt a statistical approach, examining the properties of the aerosol distribution in the trade wind cloud margin by compositing data from all cloud passes across the entire 14 flights. This allowed us to compile a large sample (Table 1) across a wide spectrum of particle sizes occurring over the tropical oceans and to characterize the mean state of the aerosol distributions in the cloud margin.

Our analyses include all aerosol data between 100 and 5000 m from all cloud boundaries for all clouds sampled on the flights, with two exceptions. 1) Aerosol data with RH > 99.5% were not included. This was done because the iterative solution to the Köhler equation (as discussed below) no longer converged as the RH approached close to 100%. Less than 1% of the total samples were removed as a result of this criterion. 2) Aerosol data from a 0.4-s period on flight RF14 and a 0.6-s period on flight RF11 were not included. These two periods had extremely anomalous high aerosol counts and may have been the result of encountering a ship plume or a close encounter with a cloud to the side of the aircraft. In either case, the data were unrepresentative of all of the remaining data from the flights.

a. Cloud boundary identification

A Commonwealth Scientific and Industrial Research Organization (CSIRO) Particle Measuring Systems (PMS) hot-wire King probe (King et al. 1978), and a Gerber Particulate Volume Monitor probe (PVM-100, Gerber 1994), located on separate wings of the C-130, measured values of liquid water content (LWC), both at 25-Hz resolution. Examination of the time series of LWC from both instruments for all the flights revealed that both probes performed well on the entrance sides of clouds. On the exit sides, the King probe had a long response time when exiting cloud (Vidaurre et al. 2011), likely because of evaporation of residual water on the probe’s hot-wire sensor, blurring the cloud edge, while the PVM-100 response was rapid, exhibiting a sharp drop in liquid water content to the clear-air threshold. For this reason, only the measurement of the PVM-100 was used for cloud-boundary identification. To match the time resolution of the particle size measurements, LWC data from the PVM-100 were downgraded to 10 Hz.

The PVM had a small zero offset during RICO that varied in the mean from 0.013 to −0.003 g m$^{-3}$ during the 14 flights included in this analysis. The zero offset of the PVM is sensitive to how well the PVM is mounted parallel to the streamlines and to what degree the collimator lens in the PVM becomes coated with some particles/drops. The minor offsets on the flights did not vary systematically, suggesting that these issues were not a problem. To account for the minor drifts in the zero offset, thresholds to determine cloud edge were automatically chosen for each flight based on the frequency distribution of the LWC for each flight. As an example of the procedure, Fig. 1 shows the histogram of LWC for flight RF04. Most of the flight was outside cloud, and the distribution with the peak near −0.005 g m$^{-3}$ represents clear conditions. Assuming that the white noise of the instrument is normally distributed, we chose a threshold at the right side of the noise distribution of Fig. 1 with the same distance to the peak as the distance between the lowest recorded LWC value and the peak value. For this example, the threshold was set at −0.003 g m$^{-3}$. A 10-Hz sample point was classified as in-cloud if its LWC value was equal to, or larger than the threshold; otherwise the sample was classified as out-of-cloud. Changing the threshold from −0.005 to +0.003 g m$^{-3}$ in this case caused an approximately 5% decrease in the amount of 10-Hz sample points flagged as cloudy and had minor effects on the frequency distribution of the cloudy sample points. As a second, much stronger precaution against including cloud particles, we excluded data within 100 m of all cloud edges as determined by the PVM-100 threshold technique. Adjacent points exceeding the LWC threshold were considered to belong to the same cloud. If the spacing between adjacent clouds was less than 200 m, data within the gap were ignored, since all points within the gap were less than 100 m from a cloud edge.

Figure 2 illustrates the sampling procedure. The flight strategy during RICO was to choose a target cloud and pass through it. After exiting the cloud, the pilot and flight scientist identified the nearest cloud that could be targeted with normal maneuvering and turn rates. The aircraft was lined up to hit the center of the new target cloud and then approached it. Each cloud the aircraft...
**Table 1.** Flight data used in study and LWC thresholds for cloud identification. The data used were from the middle 4 h of each flight during the period of cloud sampling. The first row shows the RICO flight numbers. The second and third rows are the start and end times of the cloud-sampling period. The fourth row specifies the chosen LWC thresholds. The remaining rows show the number of 10-Hz samples outside of cloud used in the analysis of the aerosol distributions. A sample point at 10 Hz during a flight was considered out of cloud only if its LWC value was smaller than the threshold determined for each flight.

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**Altitude bin (km)**

**Number of 10-Hz samples in each layer**

- [0.6–0.7) 530 35710 136 12152 15209 612 1219 2345 783 607 413 583 679 998
- [0.7–0.8) 1200 712 16088 18468 326 517 28999 13708 29854 2247 11915 3403 598 18293
- [0.8–0.9) 32739 852 749 17542 10150 31434 2758 14277 1284 43803 1169 1134 515 991
- [0.9–1.0) 646 42681 339 890 10592 17921 11689 2833 317 452 593 5358 599 18749
- [1.0–1.1) 575 1036 15953 923 13894 2419 765 13850 225 374 9419 595 1053 2760
- [1.1–1.2) 540 478 10295 28322 14437 16830 964 34197 239 331 1109 556 609 9197
- [1.2–1.3) 548 1310 364 2251 11521 424 7677 300 244 292 324 529 5414 2953
- [1.3–1.4) 701 28048 404 491 12252 16022 9474 293 249 29576 380 547 1492 816
- [1.4–1.5) 10792 0 16502 2007 222 391 369 13417 244 235 365 2274 1121 2188
- [1.5–1.6) 3701 0 329 18720 146 414 354 1319 268 302 804 5049 322 11729
- [1.6–1.7) 1342 0 116 3879 132 22854 525 9305 266 13316 5527 943 353 681
penetrated had an entry point \((\phi_1, \theta_1)\) and an exit point \((\phi_2, \theta_2)\), where \(\phi\) is latitude, and \(\theta\) is longitude. The horizontal distances across Earth’s surface along great circles from the aircraft to the exit point of cloud \(x\), and the entry point of cloud \(x + 1\) were calculated. The length of the shortest leg (e.g., leg B on Fig. 2) was stored for each clear-air sample point. We compared the distance calculated in this way with the distance calculated using aircraft speed for different time intervals when the aircraft flew straight at a constant altitude, and the differences are all within 1%. The advantage of the former method was that we could account for aircraft turns during the flight (e.g., Fig. 2). The possibility exists that an undetected cloud might be located off the track and might be nearer to the aircraft than either of the clouds sampled (e.g., cloud 3 in Fig. 2). There was no way to account for such clouds, and their possible influence represent a source of uncertainty in the interpretation of the analyses that follow.

Figure 3 shows the number of 10-Hz samples collected as a function of altitude and distance to cloud. Table 1 shows the number of 10-Hz samples contributed by each flight within each altitude bin on Fig. 3. Aircraft passes through clouds were typically made at a constant altitude for about 40–60 min. Following that, another altitude was chosen and flown for a similar time period. Over the period of the project, certain altitudes were flown more frequently than others, so the amount of data collected was greater at some altitudes than others. Also, the number of samples collected closer to clouds generally exceeded the number farther from clouds, since the aircraft did not fly out long distances after exiting a cloud before targeting the next cloud.

b. Humidity and aerosol spectra measurements

An NCAR model cross-flow Lyman-Alpha hygrometer (Buck 1976) was used to measure RH. The instrument can function adequately for absolute humidities between 0.5 and 25 g m\(^{-3}\). The baseline for the instrument tends to drift during flight thus reducing its accuracy in terms of true dewpoint values or absolute humidity. Periodic adjustments to the baseline were done by comparison with the C-130 EG&G hygrometer. With these adjustments, the accuracy could be maintained at \pm 4\% for relative humidity and \pm 0.6\degree C for dewpoint values. The response of the instrument is unimpaired by temperature or humidity extremes and measures every 2 ms, which is faster than the normal data system sampling rate (Friese et al. 1986; Schanot and Spyers-Duran 1987; Schanot 1987).

The Thermo-Systems condensation nuclei (CN) counter was used to determine total particle concentrations. The counter condenses butanol vapor on all particles between

![Fig. 1. The normalized frequency distribution of liquid water content values for flight RF04 with a bin width of 0.001 g m\(^{-3}\). The vast majority of the data collected during flights was out of cloud—the distribution to the left of zero represents these out-of-cloud samples. The frequency of cloud samples with liquid water content greater than 0.02 g m\(^{-3}\) is shown in the inset. The threshold chosen for RF04 was 0.003 g m\(^{-3}\).](https://journals.ametsoc.org/doi/abs/10.1175/JAS-D-12-0105.1?journalCode=jas)

![Fig. 2. Illustration of procedure for determining distance to cloud edge (see text).](https://journals.ametsoc.org/doi/abs/10.1175/JAS-D-12-0105.1?journalCode=jas)

![Fig. 3. Number of 10-Hz samples available to calculate spectra. The samples were binned in increments of 100 m (altitude) and 100 m (distance to cloud). The numbers in white on the right are the number of cloud penetrations. Each cloud penetration resulted in two sampling legs, one moving toward and one moving away from a cloud.](https://journals.ametsoc.org/doi/abs/10.1175/JAS-D-12-0105.1?journalCode=jas)
about 0.01- and 3.0-μm diameter so that they grow to an optically detectable size. Operating principles of the instrument can be found in Twyoh (1991). There is a 3-s delay owing to transit of air through the plumbing to the CN instrument that must be accounted for to correctly position the measured concentrations relative to cloud boundaries. The data were corrected to account for the delay during initial processing by the NCAR Research Aviation Facility.

Particle size distributions in the cloud-free air within the cloud margins were measured at 10 Hz with a PMS Passive Cavity Aerosol Spectrometer Probe (PCASP/SPP-200; Strapp et al. 1992; Snider and Petters 2008) and a PMS Forward Scattering Spectrometer Probe (FSSP/SPP-100, Nagel et al. 2007). The PCASP was located on the far-right wing mount and the FSSP on left wing pod of the C-130.

The PCASP measures particles in 30 bins. The instrument has three gain stages, where the highest gain is needed to detect the smallest particles. These gain stages also amplify electrical noise, and this noise can be a problem for the high-gain channels. For RICO, the measurements showed large variations in size bins below 0.125 μm, hence the data processing was set to use that as the minimum size, and only the upper 24 bins recorded usable data. Size bin boundaries were based on calibrations performed by NCAR Research Aviation Facility (RAF) staff before, during, and after RICO. The RAF calibrations used the industry standard approach of monodisperse-size polystyrene latex sphere particles nebulized from an aqueous suspension. The calibrations were consistent across the project. Because the composition and shape of individual aerosol particles measured by the probe in the field are unknown and variable, no corrections for particle index of refraction were applied. Based on the RAF calibrations, the PCASP during RICO measured particles with diameters between 0.125–2.75 μm.

The operating principles of the PCASP are described by Liu et al. (1992), Strapp et al. (1992), Collins et al. (2000), and Snider and Petters (2008). The C-130 PCASP has three inlet heaters. Strapp et al. (1992) concluded that because of adiabatic and diabatic warming of the sample occurring during transit to the detection optics, haze solution droplets sampled from a humid boundary layer will dry prior to arriving at the probe detection optics provided that the haze particles have initial diameters smaller than about 10 μm. Their conclusion was based on an assumption that the RH in the probe chamber is about 30%. Their study suggests that haze droplets larger than 10 μm may not achieve equilibrium size before detection. Snider and Petters (2008) argued that PCASP sizing errors can range from 10% to 30% when detecting particles with an initial wet size equal to 0.9 μm. The RH of the sample air passing through the PCASP chamber during RICO was not measured and is unknown.

In this paper, we assume that all particles regardless of size achieve equilibrium and arrive at the detector at an RH of 40%. This value is about 10% higher than that assumed by Strapp et al. (1992) to partially account for the possibility that larger particles may not have dried to their equilibrium size. We recognize that the larger particles entering the PCASP may still be improperly sized because they have not achieved equilibrium at 40% RH at the detection optics of the probe. To reduce the impact of sizing errors on interpretation of aerosol size spectra, mean particle concentrations in each spectral size bin, compiled in 100-m-wide distance bins from cloud boundaries, are normalized by mean particle concentrations in each spectral size bin in the distance bin 100–200 m from cloud boundaries. This procedure is explained further below.

The PMS FSSP/SPP-100 [see Nagel et al. (2007) for references] measured particle size spectra in 30 bins. As with the PCASP, size calibrations of the FSSP were done before, during, and after the RICO field project. The calibrations were based on uniform-size glass beads. Adjustments to the indicated size were made in order to correct for the index of refraction of glass versus water (1.33). To avoid the effects of noise in the lower size bins, RAF postprocessing zeroed out the lowest two size bins, setting the FSSP minimum usable bin to bin 3 (small bin edge at 2.35-μm diameter). Final calibrations set the usable size range at 2.35–45.75 μm. The FSSP measures particle size distributions at ambient RH. In the tropical environment, aerosol measured by the FSSP would be in the form of haze droplets. We assume herein those aerosol particles measured by the FSSP were in equilibrium at the ambient RH at the location of the measurement. Information and references concerning operating principles and accuracy of the FSSP-100 can be found in Coelho et al. (2005a,b).

Both the FSSP and PCASP concentrations can be contaminated by shattering of raindrops near the entry points of the probe orifices. Drizzle and rain sometimes were observed visually during RICO beyond cloud boundaries associated with higher stratiform clouds at the top of the trade wind layer or from the tops of sheared clouds. To eliminate raindrop contamination, we filtered all data where the two-dimensional-cloud (2DC) optical array probe exceeded 0 L⁻¹. The 2DC data were 1 Hz, so 10 samples (the 10-Hz PCASP and FSSP data) were rejected every time the 2DC exceeded 0 L⁻¹. We note that our criterion is far more stringent than the typical 1 L⁻¹ used in many studies. The filtering further removed 4.5% of the data but had little effect on the results—in general,
there was very little drizzle beyond 100 m from any of the RICO clouds sampled by the aircraft.

We were also concerned that residue from instrument wetting during cloud penetrations might lead to biases in aerosol size distributions measured after the aircraft exited a cloud, compared to aerosol spectra measured prior to entering a cloud. To test whether a bias might exist, we separately averaged the aerosol size distributions as a function of altitude and distance from cloud from all flight leg segments approaching clouds and receding from clouds. Although there were minor differences in the size distributions for entry and exit (partially due to the reduced sample size contributing to each group), no systematic differences resulted that contradict the findings we present below.

c. Köhler equation adjustments to particle spectra

To separate the effects of humidification from the effects of vertical transport and cloud processing of aerosol in the cloud margin, the FSSP particle size distribution of each 10-Hz sample was adjusted from the ambient RH value at the time and location where measurement took place to a constant RH of 95% using the Köhler equation. The PCASP particle size distribution of each 10-Hz sample was also adjusted using the Köhler equation from an assumed value of RH of 40% at the probe optics to the ambient RH at the time and location where individual spectra were measured. Inherent in the derivation of particle size distributions using this method is the assumption that equilibrium thermodynamics can be applied in the cloud margin, that is, particles, when they were observed with the instruments, were at equilibrium with their environment. We also have to assume that the particles, when measured at the instrument sensors, were at equilibrium at the humidity in the instrument chamber. There is no way to confirm that these assumptions are indeed the case, since the thermodynamic history and precise chemical composition of each sampled aerosol is unknown. Any deviation from equilibrium that was present contributes to uncertainty in the results which follow.

The Köhler equation (Köhler 1936; Fletcher 1966; Rogers and Yau 1989; Pruppacher and Klett 1997) can be expressed as

\[ S = \exp \left( \frac{2\sigma' M'}{r \rho' RT} \right) \times \left( 1 + \frac{M_w m_i}{M_s m_w} \right)^{-1}, \]

where \( \sigma' \) is the surface tension of the solution, \( M' \) is the molecular weight of the solution, \( r \) is the radius of the droplet, \( \rho' \) is the density of the solution, \( R \) is the universal gas constant, \( T \) is temperature, \( i \) is the van’t Hoff factor, \( M_w \) is the molecular weight of water, \( M_s \) is the molecular weight of the solute, \( m_i \) is the mass of the solute, and \( m_w \) is the mass of water. When the solution is in equilibrium with the surrounding air, then

\[ S = \frac{\text{RH}}{100}, \]

where RH is the relative humidity (%). To numerically compute the equilibrium radius at a given RH, a simple particle model was adopted. Based on the limited available information on the composition of individual particles from aircraft flown during RICO (Peter et al. 2008), we chose the semiempirical internal mixture model of Nilsson (1979). Following Nilsson (1979), the mass of the dry particle can be expressed as

\[ m_0 = \frac{4}{3} \pi r_0^3 \rho_0, \]

where \( \rho_0 \) is the average density of the dry particle given by \( \rho_0 = (\delta_i \rho_i + \delta_s \rho_s)^{-1} \). Here \( r_0 \) is the radius of the dry particle, and \( \delta_i \) and \( \delta_s \) are the mass fractions of the water soluble and insoluble components, respectively. The mass of the actual dissolved matter is given by

\[ m_s = \epsilon \delta_i \left( \frac{4}{3} \pi r_0^3 \rho_0 \right), \]

where \( \epsilon \) is the fraction of \( \delta_i \) that dissolves to form a solution. The mass of water is given by

<table>
<thead>
<tr>
<th>RH (%)</th>
<th>Maritime Atlantic</th>
<th>Sea salt (NaCl)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>16.85 0.01</td>
<td>— 0</td>
</tr>
<tr>
<td>30</td>
<td>14.45 0.03</td>
<td>— 0</td>
</tr>
<tr>
<td>40</td>
<td>12.00 0.05</td>
<td>— 0</td>
</tr>
<tr>
<td>50</td>
<td>9.55 0.07</td>
<td>— 0</td>
</tr>
<tr>
<td>60</td>
<td>7.10 0.11</td>
<td>— 0</td>
</tr>
<tr>
<td>70</td>
<td>4.70 0.25</td>
<td>— 0</td>
</tr>
<tr>
<td>75</td>
<td>3.60 0.50</td>
<td>— 0</td>
</tr>
<tr>
<td>80</td>
<td>3.28 0.72</td>
<td>2.926 1.00</td>
</tr>
<tr>
<td>85</td>
<td>2.80 0.87</td>
<td>2.697 1.00</td>
</tr>
<tr>
<td>90</td>
<td>2.41 0.93</td>
<td>2.429 1.00</td>
</tr>
<tr>
<td>95</td>
<td>2.13 0.97</td>
<td>2.181 1.00</td>
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<td>99</td>
<td>1.91 1.00</td>
<td>1.865 1.00</td>
</tr>
<tr>
<td>99.5</td>
<td>2.02 1.00</td>
<td>1.862 1.00</td>
</tr>
</tbody>
</table>

Table 2. The van’t Hoff factor \( i \) and the dissolved fraction \( \epsilon \) for maritime Atlantic conditions at different relative humidities (from Nilsson 1979)
\[ m_w = \frac{4}{3} \pi (r^3 - r_0^3) \rho_w, \]

where \( \rho_w \) is the density of water. The density and molecular weight of the salt solution is

\[ \rho' = \frac{m_s + m_w}{m_s \rho_s + m_w \rho_w} \quad \text{and} \quad M' = \frac{m_s + m_w}{m_s / M_s + m_w / M_w}. \]

\[ S = \frac{\text{RH}(\%)}{100} = \exp \left[ \frac{2}{\rho' \rho_0} \left( \frac{\sigma_w(T) + b \rho_0}{[(r/r_0)^3 - 1] \rho_0} \right) \left( \frac{M_s M_w ([((r/r_0)^3 - 1] \rho_w + \epsilon_s \rho_0)}{M_s \rho_w ((r/r_0)^3 - 1] + M_w \epsilon_s \rho_0} \right) \right] \times \left( 1 + \frac{M_w}{M_s} \frac{\epsilon_s \rho_0}{((r/r_0)^3 - 1] \rho_0} \right) 

To perform the calculations, numerical values of \( \sigma_w, \epsilon, \rho_s, \rho_0, b, M_s, i, \) and \( T \) are required. The temperature at the location where each particle was measured was used for \( T \). The surface tension of water \( \sigma_w \) was calculated using the relation

\[ \sigma_w(T) = \sum_{n=0}^{6} a_n T^n, \]

where the coefficients \( a_0, \ldots, a_6 \) are given by Pruppacher and Klett (1997), their Eq. (5-12). The values of the parameters \( \sigma_w, \epsilon, \rho_s, \rho_0, b, M_s, i, \) and \( T \) are dependent on the dry size of the aerosol as well as their composition. The numerical values we used are those reported in Nilsson (1979). For accumulation-mode aerosol, Nilsson assumed a composition of 80% ammonium sulfate and 20% insoluble material, either carbon or mineral dust. Measurements during RICO reported by Peter et al. (2008) support this assumption. They reported that aerosol with dry radii smaller than 0.2 \( \mu \)m were predominantly ammonium sulfate with a small insoluble component, which they assumed to be either elemental carbon or mineral dust. Nilsson (1979) reports \( \delta_s = 0.8, \rho_s = 2.0 \text{ g cm}^{-3}, \rho_0 = 2.2 \text{ g cm}^{-3}, M_s = 100 \text{ g mol}^{-1}, \) and \( b = 20 \) dyne cm\(^{-1}\) for accumulation-mode aerosol. For the coarse particle mode, we used Nilsson’s maritime Atlantic values: \( \delta_s = 0.6, \rho_s = 2.05 \text{ g cm}^{-3}, \rho_0 = 2.45 \text{ g cm}^{-3}, M_s = 80.8 \text{ g mol}^{-1}, \) and \( b = 22.5 \) dyne cm\(^{-1}\). As a test of the sensitivity of this choice, we also calculated the particle spectra assuming the coarse particle mode aerosol were pure sea salt (NaCl). For these calculations, we used Nilsson (1979) values of \( \delta_s = 1.0, \rho_s = 2.17 \text{ g cm}^{-3}, \rho_0 = 2.17 \text{ g cm}^{-3}, \)

\[ M_s = 58.45 \text{ g mol}^{-1}, \] and \( b = 27.6 \text{ dynes cm}^{-1}. \) The van’t Hoff factor and the dissolved fraction \( \epsilon \) are dependent on RH. Here we used the maritime Atlantic values and sea salt values reported by Nilsson (1979) (see Table 2).

The precise size of each particle measured by the PCASP or FSSP is unknown—only the particle concentration in each of the size bins is available. To adjust the PCASP particle size distribution of each 10-Hz sample to the ambient RH at the time and location it was observed, the following procedure was followed. First, the values of the bin edges of each size bin were passed into Eq. (9) to calculate their new values at ambient RH. Each adjusted bin retained the same particle concentration as its corresponding original bin. To compile the size distribution across many samples, we then resampled the adjusted size distribution at a constant bin width of 0.1 \( \mu \)m, assuming that particles were uniformly distributed within each of the adjusted size bins. The FSSP particle size distribution of each 10-Hz sample was adjusted using a similar procedure, but from ambient RH to a constant 95% RH. We then resampled the adjusted FSSP size distribution at a constant bin width of 1 \( \mu \)m, again assuming that particles were uniformly distributed within each of the adjusted size bins. Particle size spectra from both instruments are reported as a function of altitude (600-1700 m) and distance from cloud edge (100-5000 m) for both ambient and constant RH.

d. FSSP–PCASP comparison

Figure 4 shows a comparison of the FSSP spectra measured at ambient RH and the PCASP spectra hydrated.
to ambient RH using the Köhler equation for the complete RICO dataset integrated over the altitude range 600–1700 and 100–5000 m from cloud boundaries. The top panel assumes that coarse particle mode aerosol have Nilsson’s maritime Atlantic composition while the bottom panel assumes sea salt. The original PCASP measurements are also shown. The differences resulting from assuming maritime Atlantic versus sea salt aerosol on the hydrated PCASP spectra were very minor. In any size bin, the maximum difference in concentration never exceeded 0.015 m$^{-3}$. This suggests that a simpler Köhler model (e.g., Petters and Kreidenweis 2007) may have been adopted here, although the Köhler model that we chose represents aerosol closest to those measured by Peter et al. (2008) during RICO.

The PCASP spectra after hydration overlap the smallest five FSSP size bins (2.35–10.10 μm). The slopes of the FSSP and PCASP spectra differ, with the curves crossing within the third size bin of the FSSP. In the FSSP smallest size bin (2.35–3.90 μm), the calculated concentrations of hydrated PCASP aerosol exceeds the FSSP measured aerosol by a factor of 5.4 for maritime Atlantic and 5.2 for sea salt aerosol. The source of these differences is unknown but may be at least partially related to the evaporation time of particles at the large end of the PCSAP range and their transmission time into the optics. The faded lines give the 95% confidence intervals for the mean particle concentrations. The increasingly large spread of the confidence intervals for the largest size ranges of the hydrated PCASP concentration correspond to the area of overlap between the FSSP and PCASP. The PCASP hydrated spectra fluctuate above approximately 6 μm. There is also indirect evidence of sampling issues for the FSSP in its smallest size range in that the slope of the FSSP curve abruptly changes (becomes flatter) below 7 μm. Determining the nature of these differences is beyond the scope of this paper.

3. Effects of humidification versus transport and cloud processing

Figure 5a shows the observed average RH as a function of distance from cloud and altitude, and Fig. 5b shows the same data, but normalized by the RH in the
100–200-m distance bin at each altitude. There are three key points that we can draw from these figures. 1) The cloud “humidity halo,” the region adjacent to the cloud with elevated RH compared to the environment, extends about 1500–2000 m from the cloud edge below 1200-m altitude. Between 1200 and 1500 m, the halo appears to extend farther, to about 3000 m. This range of distances is consistent with past measurements of humidity halos in marine cumulus (e.g., Radke and Hobbs 1991; Perry and Hobbs 1996; Lu et al. 2003; Laird 2005; Twohy et al. 2009). 2) Within the cloud layer, the value of the RH within the halo increases from environmental values of about 80%–83% to cloud edge values of about 88%–92%. 3) During the sampling period, the depth of the trade wind layer varied (Davison et al. 2013a,b,c). For example, the anomalously low RH in the 1300–1400-m bin represents particular days where the aircraft sampled drier air toward the top of the trade wind layer. The halo effect at this altitude is still present, although environmental values of RH are lower.

Figure 6 shows aerosol concentrations measured by the CN counter within the cloud margin. As noted earlier, the CN counter measures particles with diameters greater than 0.01 μm. Since small aerosol normally occur in much greater concentrations, the data largely represent aerosol with diameters smaller than that sampled by the PCASP and FSSP. A sharp gradient in aerosol concentration was observed between 100 and 800 m from the cloud boundary, with aerosol concentrations decreasing by nearly an order of magnitude from near 1500–2000 cm⁻³ to a background value of 200–300 cm⁻³. There was no clear altitude dependence in these data. The aerosol concentration enhancement did not extend as far into the cloud margin as the humidity halo.

To provide a proper statistical representation of aerosol size spectra averaged over a range of altitudes, we took the following two-step approach. First, aerosol spectra were averaged within each 100-m altitude bin between 600 and 1700 m. The results were then averaged again with the mean spectra within each of the 100-m altitude bins equally weighted in the average. Figures 7 and 8 show aerosol size spectra as a function of distance from cloud, averaged over the altitude range 600–1700 m. The data are presented using a 100-m-wide bin width between 200 and 5000 m from the cloud edge. The concentrations are normalized by the concentration in the 100–200-m distance bin. No values are shown in the 100–200-m bin (in Figs. 7–14) since normalized values are all unity in that bin. As noted earlier, data from the 0–100-m distance bin were not used in the normalization to avoid potential cloud contamination and are not shown on this and subsequent figures.

To calculate the spectra in Figs. 7a and 7d, the coarse particle mode ($r > 0.2 \mu m$) was assumed to conform to Nilsson’s (1979) maritime Atlantic aerosol, while in Figs. 8a and 8d, the coarse particle mode was assumed to conform to sea salt aerosol. Figures 7a and 8a show the calculated FSSP size spectra at a constant RH of 95%, and Figs. 7b and 8b show the measured PCASP spectra at constant RH (the assumed 40% humidity at the optical detector). We can interpret these panels as illustrating the effect of vertical transport and cloud processing of
aerosol and subsequent depositing of aerosol into the cloud margin, since humidification effects, as a function of distance to cloud edge, have been largely eliminated. The right panels of Figs. 7 and 8 show the measured aerosol spectra for the FSSP (Figs. 7c, 8c) and the calculated PCASP (Figs. 7d, 8d), both at ambient RH. These panels illustrate the combined effect of all processes: vertical transport, cloud processing, and changes in aerosol size as a result of humidification within the cloud margin.

Little difference exists in the character of the normalized distributions when comparing similar panels of Figs. 7 and 8. This suggests that the variability in aerosol distributions across the cloud margin is largely insensitive to the choice of composition for coarse particle mode aerosol. Based on this finding, and the similarity of Figs. 4a and 4b, we limit the remainder of the analyses in the paper to results where the chemical composition of aerosols in the coarse particle mode corresponds to Nilsson’s (1979) maritime Atlantic values.

Examining Fig. 7a, which represents the FSSP coarse mode aerosol at 95% RH, the first obvious effect is that, as the result of the calculations, some particles increased and others decreased in diameter from their sizes at ambient RH. In general, most particles increased in size, so the lowest two size bins are not well populated. Focusing on the variability as a function of distance from cloud, we see that concentrations of coarse particle mode aerosol fall from values 100–200 m from the cloud edge to near-environmental values at about 500 m from the cloud edge, a distance about a third to a quarter of the size of the mean humidity halo. These data suggest that coarse particle mode aerosols processed by clouds are mixed outward from the cloud more slowly than the water vapor molecules produced during evaporation at the cloud boundaries. Figure 7b shows the same plot for the aerosol measured by the PCASP. In this case, we see a size-dependent effect, where smaller particles (0.2–0.5-μm mean diameter) are mixed outward farther into the humidity halo, while larger particles remain closer to the cloud boundary.

Comparing Fig. 7c, where humidity effects are included, and Fig. 7a, where they are not, there appears to be a noticeable difference in the way the spectra change as a function of distance from cloud. It is best to interpret this behavior by referring to Fig. 5. When humidity effects are not included, the enhancement of FSSP particle concentrations are limited to within about 500 m from the cloud. When humidity effects are included, the enhancement of FSSP particle concentrations extends farther into the humidity halo, particularly for the smaller particles. Comparing Figs. 7d and 7b, we see the effects of humidification on the smaller aerosol. Again, the first
obvious effect is that the particles increase substantially in size from their dry size (measured by the PCASP) to become haze droplets at ambient RH. Because the ambient RH is about 85% and all the measured particles grow substantially, nearly all of the smallest measured particles grow out of the small size bins \((0.1–0.3 \text{ m})\) leaving these bins with few if any particles. The data in these bins should be ignored since few of the originally measured particles remain at these sizes after humidification. Concentrating on how aerosol size spectra vary across the cloud margin, the aerosol effects, including humidification, are primarily again within about 800–1000 m from the cloud boundary.

Summarizing the data from these four panels, the aerosol size distributions are altered by humidification, vertical transport, and cloud processing within about 800–1000 m beyond the cloud boundary within the cloud margin. It appears that there is some size dependence on how far the aerosol enhancement extends into the cloud humidity halo. The reason for this size dependence remains a point of research.

We next determined if any altitude dependence exists to the horizontal distribution of aerosol within the cloud margin by examining the data from the six altitudes where the C-130 concentrated its sampling. Figures 9–14 show normalized aerosol spectra as a function of distance from cloud in a manner similar to Fig. 7, but for the altitude ranges 700–800, 1000–1100, 1100–1200, 1300–1400, 1400–1500, and 1600–1700 m, respectively. These were layers with the largest number of 10-Hz samples available from the flights, and layers where the largest humidity variations were observed. Figure 9 represents clouds sampled about 200–300 m above cloud base near the top of the transition layer between the surface based mixed layer and the trade wind cloud layer (Davison et al. 2013a,c). The left panels, which are at constant RH, show that aerosol concentration enhancement extends outward to about 1200–1500 m beyond the cloud edge, particularly at the smallest sizes. With the effect of humidification (right panels), the aerosols are larger, but the enhancement is again within the same distance from the cloud. In both cases, the enhancement is greatest nearest the cloud boundary. Note that during RICO, most marine cumulus were shallow, and formed at the top of the mixed layer. The clouds often were anchored along boundary layer roll circulations. Minor et al. (2011) showed that these clouds can have very long lifetimes (well over an hour), since they are continually reinforced by updrafts generated by the roll circulations. Deeper clouds penetrating toward or to the top of the trade wind boundary layer were less common. These deeper clouds tended to occur in clusters or along outflow-generated...
arcs (Rauber et al. 2007; Snodgrass et al. 2009; Zuidema et al. 2012), and their survival times at the higher altitudes were limited both because of entrainment and rainout (Reiche and Lasher-Trapp 2010; Arthur et al. 2010; Minor et al. 2011). The potentially longer average lifetimes of the smaller clouds, may have allowed the time for turbulent transport of aerosol farther into the environment to be greater, leading to the observed distributions on Fig. 8.

Figures 10–14 show aerosol size distributions for altitude ranges spanning from 1000–1100 to 1600–1700 m in format similar to Fig. 9. The left panels of the figures, which show concentrations at constant RH, show no distinct trend in aerosol concentration as a function of distance from cloud, except possibly within about 500 m of the cloud boundary. The difference between Fig. 9 and Figs. 10–14 may be attributable to the shorter lifetimes of taller clouds, so that fewer cloud-processed aerosols are mixed into the cloud environment, although this explanation must be considered speculative with the data available. When the effects of humidity are included (right panels), particle concentrations again appear enhanced within the humidity halo relative to their values farther from the cloud. Even here, the effect is not as distinct as it is at 700–800-m altitude (Fig. 9). These data together suggest that the greatest effects are in the low altitudes where most trade wind clouds are found (Zhao and Di Girolamo 2007).

4. Aerosol optical properties

As a basis for comparison to remote sensing measurements, we calculate several optical properties from the measured size distributions. To perform these calculations, we merged the FSSP and hydrated PCASP spectra appearing on Fig. 4a. PCSAP data were used for particles with diameters smaller than 6 μm, while FSSP data were used for particles with diameters larger than 6 μm. The transition size was chosen at the location where the PCASP and FSSP curves on Fig. 4 most closely coincide. The parameters calculated were the volume extinction coefficient at a wavelength of 532 nm $\alpha_{532}$, the backscatter coefficients at wavelengths of 532 and 1064 nm ($\beta_{532}$ and $\beta_{1064}$), and the color ratio $\chi = \beta_{1064}/\beta_{532}$. The parameters were calculated for each of the 10-Hz samples collected by the PCASP and FSSP at ambient RH. Mie calculations were performed using the Boheren–Huffman Mie scattering code (Bohren and Huffman 1983). We used the same datasets for refractive indices as in TD09, assuming, as they did, that the aerosol composition is ammonium sulfate for $r < 0.2$ μm, and sea salt for $r > 0.2$ μm. As in TD09, refractive indices for ammonium sulfate were taken from Andrew Lacis’ Database of Aerosol Refractive Indices (http://gacp.giss.nasa.gov/data_sets/) and for sea salt from the Optical Properties of Aerosols and Clouds
In these datasets, refractive indices are given at several fixed RH levels. We linearly interpolated them at a precision of 1% RH between 0% and 99.5% for the Mie calculations. The volume-extinction coefficient for each sample was calculated as

$$\sigma_{532} = \sum_{i=1}^{n} \pi d_i^2 Q_i N_i,$$

where \(d_i\) is the midpoint of the \(i\)th size bin, \(Q_i\) is the 532-nm extinction efficiency factor derived from the Mie calculations for diameter \(d_i\), \(N_i\) is the concentration, and \(n\) is the number of the size bins. For \(d_i < 6 \mu m\), \(N_i\) was specified as the PCASP particle concentration hydrated at ambient RH, while the FSSP particle concentration was used for \(d_i > 6 \mu m\). The backscatter coefficients were calculated in a similar manner.

Figure 15a shows the calculated average \(\beta_{532}\) as a function of distance from cloud and altitude. Figure 15b shows the same data but normalized by the average of all the \(\beta_{532}\) within 5 km of cloud edge at each altitude. The averaged color ratio \(\gamma\) is given in Fig. 15c. These figures are the same format as Fig. 1 of TD09 to facilitate comparison with CALIOP measurements from space. Note that our calculations of the backscatter coefficient are different than that measured by CALIOP—CALIOP measures the total attenuated backscatter, whereby the two-way transmittance of the atmosphere is convolved with the backscatter in the measurement. The values shown for CALIOP are median values, where the values of the backscatter coefficient are means. Means were used because the aerosol concentrations at 10 Hz had many near-zero values and thus were not normally distributed. The CALIOP color ratio is calculated from the ratio of the total attenuated backscatter at 532 and 1064 nm. From Fig. 15, the variation of \(\beta_{532}\) with altitude does not exhibit a distinguishable trend for the first 400 m away from cloud edges. Beyond 400 m, \(\beta_{532}\) increases toward lower altitudes. In the horizontal direction, \(\beta_{532}\) decreases away from cloud boundaries at all altitudes. At lower altitudes, the change in \(\beta_{532}\) away from the cloud edge is smaller compared to higher altitudes. Maximum enhancements occur at both high and low altitudes near cloud. All of these features are consistent with the observations of TD09. The altitude and distance dependency of the backscatter coefficient and color ratio shown in Fig. 15 reflects variations of the aerosol size distribution with altitude and distance to clouds, as discussed in section 3.

Figure 16 shows horizontal variability in the vertically integrated backscatter. The integrated backscatter values at 532 and 1064 nm were calculated as

$$\beta_{c} = \sum_{i=1}^{N} \beta_{i} \times 100 \text{ m},$$

where \(\beta_{i}\) is the backscatter coefficient at altitude \(i\) and distance to cloud boundary \(d\).
where $x = 532$ or $1064$ nm, $\bar{\beta}_i$ is the mean backscatter within bin $i$, and $N$ is the number of layers. Daytime observations from TD09 are also plotted as stars and plus signs in our figure for the comparison. Note that the integrated $\beta_{532}$ and $\beta_{1064}$ values decrease away from the cloud boundary as also found in TD09. Within 600 m from cloud boundaries, the integrated $\beta_{532}$ and $\beta_{1064}$ values are comparable with TD09. However, in TD09,
integrated $\beta_{532}$ and $\beta_{1064}$ values decrease outward by 31% and 42%, respectively, when comparing extinction values 0.33–2.99 km from cloud boundary, while Fig. 16 shows a decrease by a factor of 4.5 and 14 in integrated $\beta_{532}$ and $\beta_{1064}$, respectively. The difference between TD09 and our studies may be at least partially attributable to the following factors. 1) In TD09, measured backscatter is modified by the two-way transmittance of

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**Fig. 13.** As in Fig. 7, but for the altitude range 1400–1500 m.

**Fig. 14.** As in Fig. 7, but for the altitude range 1600–1700 m.
the atmosphere above the layer. This is not taken into account in our calculations. Relative to CALIOP, this would result in a sharper increase in our calculated integrated backscatter as we move toward cloud edge.

2) In TD09, backscatter values were vertically integrated between 0.5 and 2.0 km. However, aerosols between 0.5 and 0.6 km and between 1.7 and 2.0 km were excluded from our study, which would lead to lower integrated backscatter from our calculations. 3) The RICO data were collected during December 2004 and January 2005, whereas TD09 analysis of CALIOP was derived during December–February 2006–09. Strong seasonality was also reported in TD09. 4) We had a very different sampling strategy from TD09. In TD09, cloud fields were sampled through random linear transects tied to the CALIOP orbit, while in RICO we selected a target cloud, passed through it, and then once in the clear, chose another target cloud and flew toward it (Fig. 2). Tackett (2009) applied both sampling strategies to simulated 2D cloud fields. He demonstrated that the variation of backscatter with distance to clouds showed similar behavior between the two strategies within about 300 m of cloud boundaries. Beyond that range, backscatter using the random sampling strategy in TD09 changed much more slowly with distance than with the flight-sampling strategy described in this paper.

TD09 also examined the color ratio $\chi$ observed by CALIOP, a quantity that is highly sensitive to particle size. The variability of $\chi$ with altitude and distance in our study (Fig. 15c) was generally similar to TD09. In general, $\chi$ decreased away from the cloud edge. The maximum $\chi$ occurred near the cloud edge both at high and low altitudes. The value of $\chi$ was approximately unity very close to the cloud edge, indicating the presence of large particles.

Given the measured particle concentrations from the PCASP and FSSP at ambient RH, we can directly calculate the mean particle diameters $D_{532} = (\sum_d N_d)/(\sum N_d)$.

Figure 17a shows $D_{532}$ as a function of altitude and distance, and Fig. 17b shows the same data, but normalized by data in the 100–200-m distance bin. Again, $N_d$ uses the PCASP particle concentration hydrated at ambient RH for $d \leq 6 \mu m$ and FSSP particles at ambient RH for $d > 6 \mu m$. Particle diameters are larger through the humidity halo, ranging between 0.4 and 1.0 $\mu m$. Maximum diameters occur near the cloud boundary both at high and low altitudes. The horizontal variation of $D_{532}$ was larger at lower altitude. At low altitude, $D_{532}$ changed by 20%–30% from 0.3 to 3.0 km away from clouds, while at high altitudes, the change was 40%–50%. The horizontal change of $D_{532}$ averaged across all altitudes is given in Fig. 18. Note that $D_{532}$ rapidly declines from 0.53 to 0.4 $\mu m$ from near the cloud boundary to about 800 m and then decreases slightly beyond 800 m away from clouds. From 300 m to 3 km, $D_{532}$ changes by about 25%. This is consistent with the particle size changes that would reproduce the lidar-measured color ratio variation in TD09’s sensitivity studies with assumed aerosol distributions.

We also calculated the volume extinction coefficient at 532 nm $\sigma_{532}$ for each 10-Hz sample. Figure 19a shows the calculated average $\sigma_{532}$ as a function of distance from cloud and altitude, and Fig. 19b shows the same
data, but normalized by the data within 100–200 m of cloud edge at each altitude. Figure 19 has a similar pattern to Fig. 15. This is expected given $\sigma_{532}$ is proportional to $\beta_{532}$. The value of $\sigma_{532}$ gets larger at lower altitudes and increases as clouds are approached. Integrating $\sigma_{532}$ over an altitude range yields optical depth within the altitude range. The variation of aerosol optical depth (AOD) with distance to cloud averaged over the altitude range between 600 and 1700 m is given in Fig. 20. The AOD increases from 0.02 to 0.07 as clouds are approached from 800 to 100 m. Outside that range, the AOD in Fig. 20 slightly decreases with an increase in the distance to cloud boundary, ranging between 0.02 and 0.01. The ground-based sun-photometer measurements of AOD reported in Koren et al. (2007) show an increase of about 13% at 440 nm and 22% at 870 nm close to cloud edge compared to far from cloud edge, as clouds moved over the sun photometers. Factors that contribute to these differences are similar to the factors that may explain the differences between Fig. 16 and TD09. Other factors are as follows: 1) the AOD from sun photometers were derived from 15 globally distributed sites representing a wide range of aerosol conditions, 2) different cloud types were examined, 3) the AOD from sun photometers represents an altitude range from the surface to space, and 4) a passive-optical versus a microphysical approach in how cloud edge is defined.

5. Summary and conclusions

This paper examined the effect of clouds on aerosol particle concentrations and size distributions in their margins. The data came from a large number of trade wind cumulus penetrations at altitudes between 600 and 1700 m made during the Rain in Cumulus over the Ocean field campaign. Particle size distributions and particle concentrations were examined in the cloud environment as a function of altitude above cloud base and distance from cloud, both at ambient RH and adjusted to a constant RH to isolate effects of humidification from vertical transport and cloud processing. The aerosol data were then used to examine optical properties of aerosol as a function of distance and altitude within the cloud margins. Results were compared to CALIOP observations reported by Tackett and Di Girolamo (2009) over the same tropical region. The following points were drawn from these analyses.

The humidity distribution in the cloud margin exhibited the following properties: 1) the cloud “humidity halo” generally extended about 1500–2000 m from the cloud edge, consistent with past measurements; 2) the value of the RH within the halo decreased from cloud edge values of about 88%–92% to environmental values of about 80%–83%; and 3) there was no clear altitude dependence on the horizontal extent of the humidity halo, except possibly at the highest altitudes where the halo’s extent may have been extended a kilometer farther.

The aerosol distribution in the cloud margin exhibited the following properties. 1) Aerosol concentrations measured by the CN counter were higher than background values within 800 m of the cloud boundary, ranging from values of 1400–2000 cm$^{-3}$ within 100–200 m
from cloud to background values of 200–400 cm$^{-3}$ beyond 800 m from cloud. 2) There was a weak size-dependent effect on the distribution of aerosol across the cloud margin. The concentration enhancement for larger aerosol was detectable away from the cloud boundary a shorter distance than for small aerosol; the physical reason for this behavior remains a point of research. 3) Vertical transport and cloud processing of aerosol was found to contribute to the modification of aerosol size distributions within the cloud margin, particularly close to the cloud boundaries, independent of any humidification effects. 4) Enhanced aerosol concentrations extended outward farther into the humidity halo at altitudes just above cloud base compared to higher altitudes. This was attributed to the long lifetimes of the shallow clouds, which were often anchored to updraft regions of long-lived boundary layer roll circulations.

The aerosol optical properties in the cloud margin exhibited the following behavior: 1) backscatter at 532 nm did not exhibit a distinguishable trend with altitude within 400 m from cloud edges but increased toward lower altitudes beyond 400 m from the cloud edge; 2) the backscatter coefficient at 532 nm decreased away from cloud boundaries at all altitudes; 3) integrated $\beta_{532}$ and $\beta_{1064}$ values and the color ratio decreased away from the cloud boundary; 4) the mean aerosol diameter rapidly declined from 0.53 to 0.4 $\mu$m from near the cloud boundary to approximately 800 m beyond the boundary and then decreased slightly beyond that away from clouds; 5) aerosol optical depth within the humidity halo approximately exponentially increased from 0.02 to 0.07 as clouds were approached from 800 to 100 m; and 6) the trends observed agreed qualitatively with Tackett and Di Girolamo’s (2009) CALIOP measurements of trade wind cloud margin structure, although quantitative differences were noted that likely arose because of the different sampling strategies employed by the aircraft- and space-based platforms.

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