Droplet Concentration and Spectral Broadening in Southeast Pacific Stratocumulus

1 Jefferson R. Snider
1 David Leon
1 Zhien Wang

1 University of Wyoming, Department of Atmospheric Science, Laramie, WY
Abstract

Several airborne field experiments have been conducted to verify model descriptions of the initial stages of cloud condensation. Measurements of cloud condensation nuclei made below cloud, and measurements of updraft, made either in-cloud or below-cloud, are model inputs and the concentration and size distribution (spectrum) of cloud droplets are validated. Experiments conducted within cumulus clouds have yielded the most robust validations. Investigations of stratocumulus clouds are more varied, in part because of the difficulty of gauging the effects of drizzle and entrainment. Airborne lidar and radar are used here to supplement the approach used in prior studies of stratocumulus. A model verification study was conducted using data acquired during the southern hemispheric Vamos Ocean Cloud Aerosol Land Study Regional Experiment. Consistency between observed and modeled droplet concentrations was achieved. Also, predictions of the droplet size distribution’s dispersions were 70% of the measured dispersions corrected for instrument broadening. This is consistent with the conjecture that differential cloud base activation and internal mixing - the cloud processes simulated - were important drivers of true spectral broadening.
Introduction

Planetary albedo, the fraction of solar input reflected by the Earth system, is controlled by many phenomena and has a profound impact on weather and climate (Charlson et al. (2005) and references therein). Marine stratocumulus clouds are an important contributor to the planetary albedo. Also, because these clouds occur low in the atmosphere and are relatively warm, the cooling they provide by reflecting at solar wavelengths is not strongly compensated by a radiative effect occurring at infrared wavelengths (Paltridge, 1980). The global impact of marine stratocumulus is recognized because they cover a substantial fraction of the ocean, reflecting sunlight that would otherwise be absorbed, and because their albedo (Twomey, 1977) and propensity to form rain (Albrecht, 1989) can be altered by anthropogenically- and naturally-produced aerosols. Prediction of that alteration requires understanding of several phenomena: 1) processes affecting the cloud condensation nuclei (CCN) and other aerosol particles (e.g., source strength), 2) processes that lower the concentration of cloud droplets (entrainment and precipitation) and which alter the CCN activation spectrum (e.g., aerosol processing by clouds), and 3) the activation process which converts CCN to cloud droplets.

Both observational and modeling studies have probed how properties of the CCN can alter the cloud droplet number concentration ($N$) and thus impact precipitation and cloud albedo in stratocumulus systems. Typically, field observations are used to initialize an activation model and model output is compared to measurements of the actual $N$ within nearby clouds. Several schemes, based on parcel modeling, have been developed for describing $N$ as a function of the CCN spectrum and updraft (Twomey, 1959; Leaitch et al., 1986; Yum et al., 1998; Snider et al., 2003). Some activation schemes have been further developed and are now implemented as
parameterizations within multidimensional cloud models (Ghan et al., 1997; Meskhidze et al., 2005).

All of the aforementioned \( N \)-closure studies were complicated by the difficulty of measuring \( N \) simultaneous with below-cloud measurements of CCN. Brenguier et al. (2000) addressed this by deploying three aircraft, in a staggered configuration. In addition, some ground-based investigations of coastal stratocumulus have exploited remote sensing (lidar and radar) for retrieving \( N \) simultaneous with surface measurements of CCN (e.g., McComiskey et al., 2009). The multiple-aircraft approach has the disadvantage of being expensive, while ground-based coastal investigations may be unrepresentative of conditions offshore (O’Dowd et al., 2013). As we will see, airborne remote sensing, with lidar and radar, can help to alleviate some of these complications.

Because it also affects stratocumulus albedo and precipitation, the shape of the droplet size distribution or spectrum has also been the focus of observational and modeling studies (Hudson and Svensson, 1995; Hudson and Yum, 1997; Liu and Daum, 2002; Liu and Daum, 2004; Pawlowska et al., 2006). Various methods have been used predict the how the standard deviation (\( \sigma_D \)) of droplet diameter and the average droplet diameter (\( \langle D \rangle \)) vary within clouds. In parcel model simulations, values of \( \sigma_D \) and the spectral dispersion (\( \sigma_D / \langle D \rangle \)) both decrease upward from a few tens of meter above cloud base (Warner, 1969; Rogers and Yau; 1989).

Measurements of cloud droplet spectra obtained in stratiform clouds generally do not conform to these predictions (see Miles et al. (2000) for a review) and there is a body of work indicating that a component of this disagreement is broadening by instruments used to make the spectral measurements (Politovich, 1993; Brenguier and Chaumat, 2001). Using models, Cooper (1989) and Cooper et al. (2013) demonstrated how true spectral broadening can result when parcels
entering cloud base at differing updraft speeds, characterized by differing \( N \), come into contact and become mixed by turbulence. This mechanism was investigated by Politovich (1993) and Hudson et al. (2012), who studied cumulus, and by Hudson and Svensson (1995) and Hudson and Yum (1997), who studied stratocumulus. We extend the observations and analysis of the latter two publications.

Our paper is organized as follows: Sect. 2 describes the instruments; Sect. 3 describes the field project, flight patterns and our selection of 44 analysis intervals from flights conducted west of northern Chile over the southeastern Pacific Ocean; Sect. 4 describes the in-cloud and below-cloud measurements; Sect. 5 describes the remote-sensing measurements; and Sect. 6 describes the activation modeling. In the analysis section (Sect. 7), we have four objectives. First we consider observed concentrations (in-cloud measurements) and correlate these with concentrations based on activation modeling. Second, we analyze lidar-retrieved concentrations and compare these to modeled values. Third, we compare concentration probability distribution functions (PDFs) based on observations, modeling and retrieval. Fourth, we analyze spectral dispersions based on in-cloud observations, modeling and based on a theoretical relationship developed by Cooper (1989). Sect. 8 summarizes our findings.

Henceforth, the averaged concentrations are symbolized \( \langle N_{\text{obs}} \rangle \) (observed), \( \langle N_{\text{mod}} \rangle \) (modeled) and \( \langle N_{\text{ret}} \rangle \) (retrieved), and the PDF standard deviations – here referred to as widths - are symbolized \( \sigma_{\text{obs}} \) (observed), \( \sigma_{\text{mod}} \) (modeled) and \( \sigma_{\text{ret}} \) (retrieved). Tab. 1 provides detailed definitions of these quantities and makes the point that these are statistics derived from measurements. In that sense, the concentration average and the width communicate essential but incomplete information about the concentration PDF.
Five optical particle counters (OPCs), and one optical array probe (OAP), were used to measure aerosol, droplet and drizzle spectra: 1) a Passive Cavity Aerosol Spectrometer Probe (PCASP), for dried aerosol particles measured below cloud (diameter, $D$ between 0.11 and 3 $\mu$m), 2) a Forward Scattering Spectrometer Probe (F300), for haze particles measured below cloud (0.4 $\mu$m $< D < 17$ $\mu$m), 3) a 2D-C OAP for drizzle drops ($62$ $\mu$m $< D < 1590$ $\mu$m), and 4) a Cloud Droplet Probe (CDP; $2$ $\mu$m $< D < 50$ $\mu$m), an unmodified Forward Scattering Spectrometer Probe (U-F100; $3$ $\mu$m $< D < 45$ $\mu$m), and a modified Forward Scattering Spectrometer Probe (M-F100; $11$ $\mu$m $< D < 38$ $\mu$m) for cloud droplets. With the exception of research flight 4 (RF04), our analysis used data from the CDP; data from the M-F100 was used to analyze RF04. The five OPCs and the OAP were manufactured by either Droplet Measurement Technologies (DMT; Boulder, CO), or Particle Measuring Systems (PMS; Boulder, CO), and all were externally-mounted on the C-130. Laboratory calibrations of the PCASP and the F300 are described in Appendix B. Comparisons of airborne CDP and the U-F100 measurements are analyzed in Sect. 4.1. Throughout our analysis we use 1-s averaged droplet spectra and 1-s averaged droplet concentrations.

Measurements of the cumulative CCN spectrum, at relatively large values of supersaturation ($SS$), were derived using Wyoming static diffusion instrument (Snider et al., 2006; Snider et al., 2010). This instrument sampled air brought into the C-130 via a forward-facing, isokinetic, solid diffuser inlet (Shank et al., 2012). A measurement of CCN concentration is output by the Wyoming instrument every 35 s. The Wyoming CCN’s range of supersaturation extends from 0.1 to 2 % (Snider et al., 2006).
The upward-pointing Wyoming Cloud Lidar (Wang et al., 2009a; Wang et al., 2012) was used to retrieve values of the cloud droplet concentration. The lidar transmits in the near ultraviolet (\(\lambda = 0.355 \mu m\)) at a pulse repetition frequency of 20 Hz. During VOCALS, seven lidar shots were averaged, making the time between samples 0.35 s. The vertical resolution of the lidar is 3.75 m.

Remotely-sensed values of cloud top altitude \(z_{ct}\), derived using the Wyoming Cloud Radar (Wang et al., 2012; Zuidema et al., 2012), lidar determinations of cloud base altitude \(z_{cb}\), and radiometric measurements of cloud base temperature, from an upward-viewing radiometric temperature sensor (RSTT), were also used in our analysis. The RSTT uncertainty is ± 0.5 K (Zuidema et al., 2012). In addition to RSTT, values of both cloud base temperature \(T_{cb}\), and cloud base pressure \(P_{cb}\), were inferred using \(z_{cb}\) and C-130 measurements of temperature, pressure and altitude. For the latter calculations it was assumed that the layer between the aircraft and cloud base was well-mixed (i.e., \(\partial \theta / \partial z = 0\), where \(\theta\) is potential temperature).

Measurements of vertical velocity came from the C-130’s gust probe system (Brown et al., 1983). We analyze 1-s averaged measurements of vertical velocity \(w\).

3 – Flight Patterns and Data Selection

Data was collected onboard the NCAR C-130 aircraft during the Vamos Ocean Cloud Aerosol Land Study (VOCALS) Regional Experiment (Rex) (Wood et al., 2011). During VOCALS (October and November 2008), the C-130 was used to conduct 14 long-duration flights. Our focus is on level-flight measurements made below and within the southeastern Pacific stratocumulus deck. By applying criteria explained in this section, 44 analysis intervals, each with three subintervals, were selected. The subintervals are: 1) a level-flight below-cloud
segment, 2) a level-flight in-cloud segment, and 3) an above-cloud segment which was either
level-flight or a sounding. Most of the paired below-cloud and in-cloud segments (39 out of 44)
were proximate in time (< 30 min), but horizontally separated by ~100 km. The altitude of the
below-cloud segments was no larger than 170 m MSL (\( \overline{x} = 150 \pm 10 \) m, \# = 44). Eleven of 14
VOCALS flights were analyzed (Tab. 2).

Figs. 1a-b show one of the 44 analysis intervals. This example, from RF05, is used
throughout to illustrate our methods. The flight direction on this day was principally east/west
with segments of duration 10 minute below, inside and above the stratocumulus deck. Two other
flight patterns were analyzed; one with sampling similar to that in Figs. 1a-b, but with the aircraft
track directed north/south at ~75 °W (extending to 30 °S), and another with at least two 30
minute segments within the boundary layer. There are five analysis intervals in this group. Their
flight tracks were flown perpendicular to the mean boundary layer wind and subsequent tracks
were advected downwind by an amount which was several km larger than the mean advection
(Wood et al., 2011). In this case the below-cloud and in-cloud segments were picked so that they
were nearly overlapping in the advected coordinate system. For these five analysis intervals the
delay between the in-cloud and below-cloud segments varied between 45 and 75 min.

When selecting aerosol and CCN measurements from the below-cloud segments we
applied two data-acceptance criteria: 1) cloud droplet concentration < 1 cm\(^{-3}\) (Sect. 2), and 2)
drizzle drop concentration < 1 L\(^{-1}\) (Sect. 2). These were applied to eliminate bias that occurs
when hydrometeors (droplets and drops) shatter on the leading edge of a sampling inlet.
4 – In Situ Measurements

4.1 – Droplet Concentration

With the exception of RF04, droplet concentrations and droplet spectra used in this analysis came from the CDP. Because the CDP was not operational during RF04, and the U-F100 was also unavailable, measurements from the modified F100 (M-F100; Sect. 2) were used to analyze RF04.

Consistency between the CDP and the unmodified F100 (U-F100; Sect. 2) was evaluated by considering data from RF12, RF13 and RF14. The fraction of data values accepted into the comparison ($f_a$) is less than unity because the cloud liquid water content (LWC) was occasionally smaller than our in-cloud criterion ($0.02 \text{ g/m}^3$) and because some measurements were flagged invalid in the archive data. Compared to the CDP, a greater fraction of U-F100 measurements were invalidated. In Fig. 2 PDFs corresponding to the CDP (black) and U-F100 (gray) ensembles are shown. The ensemble averages ($\langle N_{obs} \rangle$), standard deviations ($\sigma_{obs}$), skewness ($sk$) and $f_a$ are presented in the legend. Both $\langle N_{obs} \rangle$ and $\sigma_{obs}$ are larger for the CDP ensemble.

Lance (2012; L12) demonstrated that concentrations reported by a CDP can be negatively biased by coincidence occurring within the probe’s sample volume. The bias is significant ($> 20\%$) for concentrations $> 270 \text{ cm}^3$. L12’s software correction was not applied in our analysis because, as can be seen in Fig. 2, the right tail of the uncorrected-CDP ensemble (black) lies to

\[\text{During RF04 the mode of the droplet spectra occurred at diameters a few \(\mu\text{m}\) larger than 11 \(\mu\text{m}\). Compared to flights that encountered droplet concentrations larger than those observed during RF04 (39 to 167 \(\text{cm}^3\); Tab. 2), when the CDP mode size was a few \(\mu\text{m}\) smaller than 11 \(\mu\text{m}\), there is less of a concern that the M-F100 undercounted during RF04.}\]
the right of the U-F100 ensemble (gray), and because we do not expect the latter to be
significantly biased by coincidence (Baumgardner et al., 1985), even during the RF12 analysis
interval associated with the third largest concentration in our data set (i.e., 265 cm$^3$ during the
first RF12 analysis interval (Tab. 2)). Concentrations can also be unbiased by restricting the
amount of scattered light reaching the CDP’s particle sizing detector (L12); however, that
correction was not available during VOCALS.

The in-cloud averaging times and the 44 in-cloud-averaged concentrations ($\langle N_{obs} \rangle$) and
concentration standard deviations ($\sigma_{obs}$) are presented in Tab. 2. These statistics were calculated
using 1-s averaged measurements associated with LWC > 0.02 g/m$^3$.

### 4.2 – Droplet Spectrum Geometric Standard Deviation

In-cloud droplet measurements were used to substantiate the assumption, in the
$N$ retrieval (Sect. 5.2), that the value we chose for the geometric standard deviation of the
droplet spectrum ($\sigma_g = 1.3$) is representative of the investigated clouds. Values of $\sigma_g$ were
derived using 1-s averaged spectra and a formula in TSI (2012). In Fig. 3 we present three $\sigma_g$
ensembles. The black (CDP) and gray (U-F100) ensembles contain values of $\sigma_g$ derived for in-
cloud segments with both the CDP and U-F100 detecting droplets ≥ 3 µm (RF12, RF13 and
RF14). In orange is the overall ensemble, consisting of the 44 in-cloud segments from 11 flights,
and with the M-F100 substituted for the CDP in the RF04.

The overall average is $\langle \sigma_g \rangle = 1.3$. It is also apparent that the CDP and U-F100
ensembles, corresponding to the RF12/RF13/RF14 subset, have consistent standard deviations
and reasonably consistent averages. Finally, we see that $1.1 < \sigma_g < 1.5$ encompasses most of the
range of the overall ensemble; consistent with the findings of Gerber (1996) and vanZanten et al. (2005).

4.3 – Vertical Velocity

The example below-cloud and in-cloud vertical velocity series, and the vertical velocity PDFs, are plotted in Figs. 4a-c. While it is apparent that the in-cloud PDF is broader (Fig. 4c), this broadening is most evident for downdrafts ( \( w < 0 \) ). It is the updraft portion of the below-cloud PDF(\( w \)) that we use to derive \( N_{mod} \) with a parcel model. This is discussed in Sect. 6.

The below-cloud averaging times and the below-cloud-averaged vertical velocities (\( \langle w \rangle \)) and standard deviations (\( \sigma_w \)) are presented in Tab. 2. Our median \( \sigma_w \) (0.4 m/s) is consistent with previous measurements made below marine stratocumulus clouds (Guibert et al., 2003; Hudson and Nobel, 2014); also, our median \( \langle w \rangle \) (0.05 m/s) is smaller than reported by Guibert et al. (2003) (0.1 m/s). No below-cloud values of \( \langle w \rangle \) were reported in Hudson and Nobel (2014).

4.4 – CCN Activation Spectra

By way of the example shown in Figs. 5a-b, and determinations of aerosol hygroscopicity (Appendix A), we now describe how measurements from the CCN and PCASP were combined into activation spectra. Our formulation of aerosol hygroscopicity, applied both in Appendix A and in the parcel model, is described using the kappa-Köhler formula of Petters and Kreidenweis (2007; their Eqn. 11). Consistent with results shown in Fig. A2b, the value \( \kappa = 0.74 \) was accepted as the project average and was used in the parcel model to describe the Köhler curves of particles with dry diameters \( > 0.11 \) \( \mu \)m. These are the particles sized and counted by the PCASP (Tab. A1). For smaller particles we assume the value \( \kappa = 0.37 \). This is consistent with a source of CCN coming from the downward mixing across the boundary layer inversion and with the fact that a
subset of the entrained free-tropospheric particles – those with critical supersaturations larger than 0.3 % - are characterized by $\kappa < 0.5$ (Wood et al., 2012; their Figure 1a). Köhler theory, with the PCASP sizing provided in Tab. A1, reveals that the range of supersaturation accessed by the PCASP (assuming $\kappa = 0.74$) is between 0.001 to 0.11 %.

Those portions of the example spectrum derived from the PCASP ($SS \leq 0.11 \%$), and the CCN ($SS > 0.11 \%$), are delineated in both Fig. 5a and Fig. 5b. We evaluated the $SS > 0.11 \%$ portion of the spectrum in two-steps. First, we fitted the Wyoming CCN measurements to a function of the form $n(SS) = C \cdot SS^k$ (Fig. 5a), and second, we discretized the $n(SS)$ function into 40 critical $SS$ classes. The latter are discernible as “steps” in the upper-right of Fig. 5b. The fitted values of $C$ and $k$ are presented in Fig. 5a, for the example, and for each of the 44 analysis intervals in Tab. 2. The spectrum at $SS \leq 0.11 \%$ is based on the below-cloud PCASP measurements, the PCASP size calibration (Appendix A), and the kappa-Köhler formula with $\kappa = 0.74$. The lower left of Fig. 5b shows the 30 critical $SS$ classes derived using the 30 optical size channels of the PCASP (Tab. A1).

Activation spectra, from the PCASP and CCN measurements made during the below-cloud segment, and droplet concentrations from the companion in-cloud segments ($\langle N_{obs} \rangle$, Tab. 2), were used to derive effective supersaturations ($SS_{eff}$; Hudson, 1984). Assuming that drizzle and entrainment do not effect $\langle N_{obs} \rangle$, the $SS_{eff}$ represents the maximum supersaturation reached within a representative cloud updraft. Since most of our $SS_{eff}$ values are smaller than 0.1 % (Tab. 3), and at these supersaturations the spectra are steeper compared to the situation at $> 0.1 \%$, where the spectrum is shallower (Fig. 5b; also see Hudson and Nobel, 2014), there is
consequence for the range of concentrations predicted by the activation model. This is discussed in Sect. 7.5.

Our median $SS_{eff}$ (0.06%; Tab. 3) is bracketed by the set of median values reported by Yum and Hudson (2002) for the FIRE stratocumulus experiment and the ASTEX experiment. The FIRE and ASTEX measurements were made over summertime subtropical oceans, but the ASTEX measurements were divided into periods affected by maritime and continental airmasses. Specific $SS_{eff}$ values in Yum and Hudson (2002) are 0.08% (FIRE), 0.31% (ASTEX maritime) and 0.04% (ASTEX continental). Although various representations of droplet concentration can be used to evaluate $SS_{eff}$, here the basis is in-cloud averages. Sect. 7.2 describes the continental influence evident in our analysis of the VOCALS CCN and droplet measurements.

5 – Vertical Profiling

Fig. 6 presents remotely-sensed cloud properties acquired during the example below-cloud segment. For a cloud forming within a well-mixed boundary layer a variation of cloud base altitude should be mirrored by a variation of cloud base temperature. This is evident in Figs. 6a-b but not so clearly in the case of the RSTT sequence. Obscuration of the expected correlation results because the RSTT’s determination of cloud base temperature is affected by decreased downwelling longwave flux during those parts of the sequence with thin or broken cloud. Examples of this are evident as RSTT minima at 10:29:10, 10:32:10 and 10:33:30 in Fig. 6b. However, in most portions of the sequence, RSTT and $T_{cb}$ are strongly correlated. With the exception of the three RSTT minima and the lidar’s detection of low cloud (scud) at 10:28:30, the temperature sequence reveals a cold-shift of $T_{cb}$ relative to RSTT (cold shift $\equiv RSTT - T_{cb}$). The median cold shift is +0.9 °C in Fig. 6b.
We also analyzed measurements acquired during climbs executed at the end of the below cloud segments. For the climb subsequent to the segment shown in Fig. 6, the upward $\theta$ increase is $+0.2$ K km$^{-1}$. For the 44 climbs, one corresponding to each of the analysis intervals, the median $\theta$ increase is $+0.4$ K km$^{-1}$. Therefore, we can say that a fraction of the cold shift might be attributable to thermodynamic stability within the sub-cloud layer. The rest of the cold shift is likely due to positive bias in RSTT. We do not probe the suspected bias in RSTT, but we do evaluate uncertainty in cloud base temperature, due to either measurement error or to departure from the well-mixed assumption (Sect. 2), and account for how that error propagates into the retrieval (Sect. 5.3).

Additionally, LWC measurements acquired during the in-cloud portions of the climbs were compared to predictions of an adiabatic cloud model. The model is essentially that used by prior investigators (e.g., Gerber, 1996) and is explained later in Sect. 5.2. For each climb an adiabatic liquid water path (LWP), a CDP-based LWP, or a M-F100-based LWP, were derived. These were integrated over the lower 150 m of the in-cloud portions of the climb and were used to evaluate departures from the adiabatic model. Because 12 of the 44 ascents passed through a break in the cloud, ended at an altitude less than 150 m above cloud base, or encountered cloud thinner than 150 m, only 32 departures are available for further analysis. Of these, 23 had LWP values within $\pm 50\%$ of the adiabatic model. Of the nine with absolute departures greater than 50 $\%$, all had a measured LWP greater than the adiabatic LWP.

5.1 - Lidar-derived Extinction Profile and Cloud Base

Our retrieval of $N_{ret}$ begins with the lidar’s measurement of a vertical profile of the attenuated backscatter coefficient. The analysis has two steps: 1) inversion of the profile of attenuated backscatter to a profile of the extinction coefficient (Klett, 1981), and 2)
determination of $N$. In the first step, it was assumed that the scatterers are large relative to the wavelength of the lidar ($\lambda = 0.355 \, \mu m$), and thus the extinction-to-backscatter ratio was set to the constant value expected for a population cloud droplets (O’Connor et al., 2004). In addition, a relationship between the lidar-measured depolarization (Wang et al., 2009a) and multiple scattering (Hu et al., 2007) was applied to correct for the effect of multiple scattering on the lidar-derived extinction coefficient. The inversion, or first step, also requires an estimate of the extinction coefficient ($\beta$) at a point in the cloud that produces diminished, but not negligible, returned power. This particular $\beta$ defines an upper boundary condition (Klett, 1981). Once the extinction profile is evaluated, in the first step, the cloud base altitude ($z_{cb}$) can be derived; the basis for this is described in Platt et al. (1994) and Wang and Sassen (2001). Reliable extinction values are available for altitudes below where the upper boundary condition is chosen ($z_{cb} + 150 \, m$).

**5.2 - Droplet Concentration Retrieval**

The focus of the second step is a layer extending upward from 20 m above $z_{cb}$. For this layer we evaluated many theoretical extinction profiles, and compared these to the lidar-derived extinction profile, and from these comparisons we inferred the optimal $N$. The theoretical extinction profiles are dependent on two assumptions: 1) the LWC profile is described by an adiabatic model (Albrecht et al., 1990), and 2) the droplet spectrum is a lognormal with constant $N$ and constant geometric standard deviation ($\sigma_g = 1.3$; Sect. 4.2). Because of the first assumption we refer to any one of the theoretical extinction profiles as an “adiabatic extinction.” The equation we use to describe the adiabatic LWC is

$$LWC(z) = \Gamma \cdot (z - z_{cb}) \quad \text{for} \quad z_{cb} \leq z \leq z_{ct} \quad (1)$$
where \( z \) is altitude and \( \Gamma \) is the adiabatic LWC lapse rate\(^2\).

The adiabatic extinctions were evaluated by independently incrementing two properties:

1) \( T_{cb} \) (varied within \( \pm 0.5 \) K of the \( T_{cb} \) described in Sect. 2), and 2) \( z_{cb} \) (varied within \( \pm 20 \) m of the \( z_{cb} \) described in Sect. 2). These properties, and the adiabatic model, define an adiabatic profile of LWC. Another equation (Frisch et al., 1995; their Eqn. 2) was used to relate \( N, \sigma_g \), and the lognormal’s geometric mean diameter (\( D_g \); TSI, 2012), to an adiabatic extinction.

\[
\beta = (\pi/2) \cdot N \cdot D_g^2 \cdot e^{2(ln \sigma_g)^2}. \tag{2}
\]

Because LWC is a function of altitude (Eqn. 1), both \( D_g \) and the adiabatic extinction coefficient \( \beta \) (Eqn. 2) are altitude dependent.

The lidar-derived profile was compared to the adiabatic extinctions, over a range that extended from \( z_{cb} + 20 \) m to approximately \( z_{cb} + 150 \) m, and a relative error statistic was formulated for each of the comparisons

\[
RE = \frac{1}{n} \left( \frac{\left( \beta_{i,\ell} - \beta_i \right)^2}{\beta_{i,\ell}^2} \right)^2 \tag{3}
\]

Here \( n \) is the number of lidar range gates within a profile, \( \beta_{i,\ell} \) is an element of the lidar-derived extinction profile, and \( \beta_i \) is an element of an adiabatic profile. The set \( \{T_{cb}, z_{cb}\} \) associated with the \( N \) that minimizes \( RE \) was identified as the retrieved cloud droplet concentration \( N_{\text{ret}} \).

\(^2\) Evaluated using Eqn. 1, in Albrecht et al. (1990), multiplied by the density of air. The temperature- and pressure-dependent \( \Gamma \) was evaluated at a state defined by the \( T_{cb} \) and \( P_{cb} \) values described in Sect. 2.
Fig. 7 illustrates the adiabatic calculation for synthetic clouds with the same LWC lapse rate (2 g m\(^{-3}\) km\(^{-1}\)) and two values of N and \(\sigma_g\). The basis for the retrieval is apparent in the sensitivity of the extinction to N. Also evident is sensitivity, albeit weaker, to \(\sigma_g\). We quantify the \(N - \sigma_g\) and \(N - \Gamma\) sensitivities in the next section.

Fig. 6 shows gaps in the radar determination of \(z_{ct}\) and in the lidar-derived quantities (i.e., \(z_{cb}\), \(T_{cb}\) and \(N_{ret}\)). Gaps in the radar sequence (i.e., between 10:32 and 10:34) result because the radar-retrieved echo was less than the minimum detectable by the upward-pointing beam of the Wyoming Cloud Radar (-18 dBZ at 1 km). Also, gaps in the lidar-derived properties (e.g., at ~10:38:30) result because we discarded lidar retrievals associated with \(RE > 0.01\) (Eqn. 3). Although not relevant to the sequence in Fig. 6, some of the below-cloud segments have values of \(N_{ret}\) coincident with RSTT colder than -20 °C. These were also discarded.

5.3 – Error Analysis

As we described in the previous section, theoretical profiles of LWC and \(\beta\), and a lidar-derived extinction profile, are the elements we use to retrieve \(N_{ret}\). A foundation of the calculation is the \(T_{cb}\) - and \(P_{cb}\)-dependent value of \(\Gamma\) (Eqn. 1). For the following error analysis we fix \(P_{cb}\) at a value representative of the southeast Pacific stratocumulus (900 hPa). This is justified because \(\Gamma\) is more sensitive to \(T_{cb}\), compared to \(P_{cb}\) (Tab. 4). With \(P_{cb}\) approximated as a constant, we now demonstrate how shifts in \(T_{cb}\) and \(\sigma_g\) affect \(N_{ret}\). For these objectives we adopt the following equation (Young, 1962; their Equation 2.9)

\[
\frac{N - N_o}{N_o} = (\Gamma - \Gamma_o) \left( \frac{1}{N} \frac{\partial N}{\partial \Gamma} \right)_{\Gamma_o, \sigma_g, o} + \left( \sigma_g - \sigma_{g,o} \right) \left( \frac{1}{N} \frac{\partial N}{\partial \sigma_g} \right)_{\Gamma_o, \sigma_{g,o}}
\]  

(4a)
Here $N$ is a perturbed concentration and $N_o$ is the concentration retrieved using default settings (i.e., $\sigma_{g,o} = 1.3$ and $\Gamma_o$ evaluated using the lidar-derived $T_{cb}$). The normalized derivatives in Eqn. 4a are useful for quantifying how the concentration is altered, in a relative sense, when a parameter different from the default setting is used in the retrieval. The two derivatives were evaluated analytically and we confirmed those findings numerically. The result is provided in Eqns. 4b-c.

\[
\left( \frac{1}{N} \frac{\partial N}{\partial \Gamma} \right)_{\Gamma_o, \sigma_{g,o}, \sigma_{g,o}} = \frac{-2}{\Gamma_o} \quad \text{(4b)}
\]

\[
\left( \frac{1}{N} \frac{\partial N}{\partial \sigma} \right)_{\Gamma_o, \sigma_{g,o}, \sigma_{g,o}} = \frac{6 \cdot \ln(\sigma_{g,o})}{\sigma_{g,o}} \quad \text{(4c)}
\]

First we investigate the lidar-derived values of $T_{cb}$, their departure from RSTT (Fig. 6b), and how that departure affects the retrieved $N$. In this analysis, we indicate the default setting with subscript “o” (i.e., $T_o = T_{cb}$) and we do not subscript the perturbed setting (i.e., $T = \text{RSTT}$). Fig. 8a portrays the point-by-point temperature differences as a distribution. The $T - T_o$ values used to construct the gray distribution come from the example below-cloud segment (Fig. 6b). Also shown is the distribution aggregated from the 44 below-cloud segments. We see that the modes of these distributions are at +0.9 °C (example) and at +0.3 °C (overall), and we note that the first of these central values was discussed in Sect. 5. Also evident (Fig. 8a) is a broad left tail in the overall $T - T_o$ distribution; this is due to the radiometric sensor’s large underestimate of the cloud base temperature when it views thin or broken cloud (Sect. 5.2). Fig. 8b has the distribution of $\Gamma - \Gamma_o$. The pattern seen here is consistent with that in Fig. 8a.
Fig. 8c shows the distribution of the relative $N$ error. This was derived using using Eqn. 4a, with the $\sigma_g$ derivative set to zero, Eqn. 4b, and the distributions in Fig. 8b. The modes of these distributions (Fig. 8c) tell us that the cloud base temperature uncertainty translates to an error in $N$ which is -8% (example) and -5% (overall). We note that smaller values of $N$ would have resulted had the generally larger values of RSTT been applied in the retrieval. This makes physical sense because a warmer cloud base means that the adiabatic LWC is increased, and therefore an $N$ smaller than $N_o$ must be applied to match the adiabatic extinction to the lidar-derived extinction.

The question of a systematic $\sigma_g$ bias is relevant because the $\sigma_g$ assessment (Sect. 4.2) was based on in-cloud measurements made ~300 m above cloud base (Twohy et al., 2013). While this altitude was useful for documenting cloud properties, it may have produced a $\sigma_g$ unrepresentative of the lower half of the cloud probed by the lidar. Parcel model predictions of $\sigma_D$ with ascent upward from cloud base reinforce this suspicion (Rogers and Yau, 1989; their Figure 7.4). Relative concentration errors, calculated using Eqn. 4a, with the $\Gamma$ derivative set to zero, and using Eqn. 4c, are presented in Tab. 5. The value $\sigma_g = 1.05$ (Tab. 5) is representative of output from our parcel model evaluated at $(z - z_{cb}) = 85$ m. We note that in the limit of closed parcel ascent, with $\sigma_g = 1.05$, the effect on $N_{ret}$ is a 13% underestimate. The physical reason for this is that a decreased $\sigma_g$ means that an $N$ smaller than $N_o$ must be applied to match a lidar-derived extinction. An example of the positive $N - \sigma_g$ correlation is shown graphically in Fig. 7, where two hypothetical lidar-derived states (red triangles) are matched by red curves. On the left, the curve is defined by $N_o = 21.5$ cm$^{-3}$ and $\sigma_{g,o} = 1.3$ and this is seen to intersect a
state which can also be matched by \( N = 20. \text{ cm}^{-3} \) and \( \sigma_g = 1.2 \). On the right, the curve is defined by \( N_o = 212 \text{ cm}^{-3} \) and \( \sigma_{g,o} = 1.3 \) and this is seen to intersect a state which can also be matched by \( N = 200 \text{ cm}^{-3} \) and \( \sigma_g = 1.2 \).

6 – Parcel Model Concentration

Modeled concentrations (\( N_{mod} \)) were derived using the parcel model described in Snider et al. (2003). Different from the description in Snider et al. (2003), the model we employed describes the relative humidity (\( RH \)) over haze particles and cloud droplets in terms of a temperature-independent and composition-independent formula, for the Kelvin effect, and in terms of a dimensionless parameter known as “kappa” - symbolized \( \kappa \) - for the solute effect (Petters and Kreidenweis, 2007). We refer to the combination of the Kelvin and solute effects as the Köhler model. The Kelvin term was formulated as in Snider et al. (2010).

The model’s initial thermodynamic state was evaluated by extrapolating the below-cloud averages of \( T_{cb} \) and \( P_{cb} \) (Sect. 2), at constant specific humidity and constant specific entropy, to \( RH = 95 \% \) (Iribarne and Godson, 1981; Chapter 7). Haze particle diameters at \( RH = 95 \% \) were derived using the Köhler model and using our description of \( \kappa \) (Sect. 4.4). The parcel model was also initialized with below-cloud-averaged CCN activation spectra; to account for the hydrostatic decrease of CCN concentration with altitude, these were scaled by the ratio of the air density at the model’s initial state divided by the below-cloud-segment air density. Non-hydrostatic changes in PCASP concentration, detected below cloud during some of the C-130 climbs (not shown here) were not accounted for. The parcel model was used to evaluate sets of droplet concentration (\( N(\caret) \)), and droplet spectra, at 200 m above cloud base. The updrafts used in these simulations ranged from 0.05 to 3.0 m/s in 0.05 m/s increments.
The average concentration was evaluated as

$$\langle N_{\text{mod}} \rangle = \sum N(w_i) \cdot PDF(w_i) \cdot \Delta w.$$  (5a)

Here $PDF(w_i) \cdot \Delta w$ is the fraction of updraft measurements in an interval $\Delta w$ centered at $w_i$.

Eqn. 5a represents the average as a convolution of two opposing effects: 1) the increase of $N$ with updraft, and 2) the smaller frequency of occurrence of large updrafts relative to small updrafts (Fig. 4c). We refer to $\langle N_{\text{mod}} \rangle$ as an unweighted average. In addition, an updraft-weighted average was evaluated

$$\langle N_{\text{mod},w} \rangle = \frac{1}{\sum w_i \cdot PDF(w_i) \cdot \Delta w} \cdot \sum N(w_i) \cdot w_i \cdot PDF(w_i) \cdot \Delta w.$$  (5b)

Some consideration of the relevance of Eqns. 5a-b is needed. Provided air parcels are unaccelerated, Eqn. 5a is the expectation for sampling of measured concentrations, or model-derived concentrations, within upward moving air a few tens of meter above cloud base (Meskhidze et al., 2005). Analogously, we see that the numerator of the rhs of Eqn. 5b can be interpreted as the flux of droplets entering a cloud model grid box. Hence, the values of $\langle N_{\text{mod},w} \rangle$ (Tab. 2) and the vertical velocity $PDF$ (e.g., Fig. 4c) can be used to validate a droplet source function within a model scheme (Ghan et al., 1997; Meskhidze et al., 2005).

The next two equations define the standard deviations (a.k.a, widths (Sect. 1)) of the unweighted and updraft-weighted concentration $PDF$s.

$$\sigma_{\text{mod}} = \left( \frac{1}{\sum (N(w_i) - \langle N_{\text{mod}} \rangle)^2 \cdot PDF(w_i) \cdot \Delta w} \right)^{1/2}$$  (5c)

$$\sigma_{\text{mod},w} = \left( \frac{1}{\sum w_i \cdot PDF(w_i) \cdot \Delta w} \cdot \sum (N(w_i) - \langle N_{\text{mod},w} \rangle)^2 \cdot w_i \cdot PDF(w_i) \cdot \Delta w \right)^{1/2}$$  (5d)
The 44 model-averaged concentrations, derived using Eqns. 5a-b, and the widths ($\sigma_{mod}$ and $\sigma_{modw}$), from Eqns. 5c-d, are presented in the final four columns of Tab. 2.

7 - Results

7.1 - Concentration Probability Distribution Functions

Averaged concentrations $\langle N_{obs} \rangle$ (Sect. 4.1), $\langle N_{ret} \rangle$ (Sect. 5.2), $\langle N_{mod} \rangle$ and $\langle N_{mod,w} \rangle$ (Sect. 6) are available for each method (retrieved, observed, unweighted model and updraft-weighted model) and for each analysis interval. The four PDFs, for the example interval, are shown in Figs. 9a-b. The observed and retrieved PDFs are presented in Fig. 9a. Values aggregated into these ensembles come from the appropriate segment of the analysis interval; e.g., the PDF($N_{obs}$) was derived by the aggregating measurements from the in-cloud segment. The locations of the in-cloud and below-cloud segments, for the example interval, are shown in Figs. 1a-1b and the $N_{ret}$ sequence is plotted in Fig. 6c.

The two model-based PDFs (unweighted and updraft-weighted) are presented in Fig. 9b. The unweighted PDF was binned into the same regularly spaced categories used for the observed and retrieved values, but the binning used for PDF($N_{mod,w}$) is dependent on the updraft increment (0.05 m/s; Sect. 6) and is therefore somewhat different. The averages are $\langle N_{ret} \rangle = 77$ cm$^{-3}$, $\langle N_{obs} \rangle = 90$ cm$^{-3}$, $\langle N_{mod} \rangle = 105$ cm$^{-3}$ and $\langle N_{mod,w} \rangle = 129$ cm$^{-3}$. The widths are different as well. In the bottom panel it is evident that updraft-weighting attenuates the left tail of the PDF, making that width the smallest of the four presented, while in the top panel a larger width is evident for the retrieval-based PDF. The widths are ranked in the following...
order: observation ($\sigma_{obs} = 17$ cm$^{-3}$), updraft-weighted model ($\sigma_{mod,w} = 23$ cm$^{-3}$), unweighted model ($\sigma_{mod} = 33$ cm$^{-3}$) and retrieval ($\sigma_{ret} = 50$ cm$^{-3}$).

7.2 – Longitudinal Dependence

Systematic variation of droplet concentration within the southeast Pacific stratocumulus sheet is documented in published analyses of the VOCALS data set and in work that preceded the campaign. The longitudinal gradients evident in Figs. 10a-d, with smaller values of concentration to the west, are consistent with the southerly low-level airflow along the Chilean coast and with the continental aerosols carried by that flow (e.g., Kuang and Yung, 2000; Wood et al., 2008; Hawkins et al., 2010; Wood et al., 2012; Twohy et al., 2013). In addition, a comparison of Figs. 10a-b establishes that retrieval-based averages are, with some exceptions, smaller than observation-based averages. It is also generally true that $\langle N_{obs} \rangle < \langle N_{mod} \rangle$ and that $\langle N_{obs} \rangle < \langle N_{mod,w} \rangle$.

7.3 – Decoupling

Within a stratocumulus-topped boundary layer it is common for a thermodynamically-defined lifted condensation level (LCL) to occur a few hundred meters below a lidar-derived cloud base (Bohren and Albrecht, 1998; their Figure 6.1). In their analysis of C-130 data from VOCALS, Jones et al. (2011) defined a decoupled boundary layer with the criterion $z_{cb} - \text{LCL} > 150$ m and surmised that two processes were responsible for the decoupling: 1) the entrainment of above-cloud air into the boundary layer, and 2) drizzle. The concept here is that entrainment and drizzle tend to increase cloud base; this is counter to the common assumption that entrainment and drizzle result in sub-adiabatic LWC but not necessarily an increased $z_{cb}$. In
what follows, we interpret the decoupling microphysically, and derive a correction factor we
subsequently apply in the comparisons of cloud droplet concentrations.

The basis for our correction is our two independent assessments of cloud base altitude.
Consider the example below-cloud segment (Fig. 6) and the lidar’s measurement of \( z_{cb} \) during
that segment. When combined with a radar-derived cloud-top altitude, a cloud thickness (\( H_1 \))
can be derived. Also, the lidar-derived \( z_{cb} \), combined with C-130 measurements of temperature,
pressure and altitude, can be used to derive an adiabatic LWC lapse rate (\( \Gamma_1 \)). Alternatively,
measurements of temperature, dew point and pressure can be used to derive an LCL and the
adiabatic LWC lapse rate at the LCL (\( \Gamma_2 \)). When the LCL information is combined with the
radar-derived cloud-top altitude, a second cloud thickness (\( H_2 \)) can also be derived. To derive
the LCL we used dew points measured by the C-130’s chilled mirror hygrometer; these were
increased by 0.8 °C to account for a known bias (Bretherton et al., 2010; Ziudema et al., 2012).

We interpret the microphysical impact of the decoupling in the following way. First,
using definitions made in the previous paragraph we denote a lidar-derived and a
thermodynamically-derived value of cloud-top LWC as \( \Gamma_1 H_1 \) and \( \Gamma_2 H_2 \), respectively. Then we
ratio these LWC values and equate the ratio to a factor we define as the decoupling factor (\( DF \))
\[
DF = \frac{\Gamma_1 H_1}{\Gamma_2 H_2} = \frac{N}{N_A}
\]  
(6)
In Eqn. 6 we see the droplet concentration (\( N \)) resulting from a combination of processes
(activation, entrainment, and drizzle) and the value (\( N_A \)) solely due to activation. Implicit in Eqn.
6 is the assumption that entrainment evaporates cloud droplets in proportion to the mass of liquid
evaporated – as is the case in the inhomogeneous limit (Devenish et al. (2012) and references
therein) – and that drizzle is an effective removal process for cloud droplets (collection
efficiency > 0.2). According to laboratory measurements, and theory, this occurs when droplet diameter exceeds ~10 µm and these are intermingled with drizzle drops with diameter larger than ~100 µm (Rogers and Yau, 1989; their Table 8.2).

Averaged values of the $DF$ were derived for 38 of the 44 below-cloud segments. The reason for missing averages is that $z_{ct}$ is not available because the radar reflectivity was less than the minimum detectable by the upward-pointing beam of the Wyoming Cloud Radar (-18 dBZ at 1 km) during six below-cloud segments. The below-cloud averaged $DF$ values range from 0.1 to 0.9 ($\bar{x} = 0.6 \pm 0.2$, # = 38) and are thus constrained between zero and one in a manner consistent with the definition provided in Eqn. 6. Also, two requisite characteristics of the $DF$ are evident. In general, the $DF$ was relatively large ($DF \sim 1$) in regions where the lidar profiles show cumuli penetrating into stratocumulus, and it was relatively small ($DF \sim 0$) in regions with higher-based stratocumulus. Examples of both cloud types are evident in Ziudema et al. (2012) (their Figure 8).

Fig. 11 demonstrates that the modeled concentrations are approximately 30 % larger than the observed values (black circles and black fit line of the form $Y = a \cdot X$). The figure also shows that the $DF$ decreases the modeled values (gray circles) and that this decrease makes the fit line (gray) statistically indistinguishable from unity indicating good agreement between the $DF$-adjusted model ($DF \cdot \langle N_{mod} \rangle$) and observed values ($\langle N_{obs} \rangle$). Our statistical inference holds true regardless of whether values on the ordinate or abscissa are used to evaluate departures from the fit line and the margin of error associated with the fitted coefficient (Havilcek and Crain, 1988). In a regression of the retrieved concentrations ($\langle N_{ret} \rangle$) plotted versus scaled model concentrations ($DF \cdot \langle N_{mod} \rangle$) the fitted coefficient is also indistinguishable.
from unity (not shown); however, this is only true when the abscissa values (i.e., $DF \cdot \langle N_{mod} \rangle$) are used to evaluate the evaluate departures from the fit line and the margin of error associated with the fitted coefficient. More variability is also apparent; quantitatively this is consistent with the fact that the square of the Pearson correlation coefficient ($r^2$) for the $\langle N_{ret} \rangle$ versus $DF \cdot \langle N_{mod} \rangle$ regression (not shown) is almost a factor of two smaller than the $r^2$ in Fig. 11. Hence, we cannot be categorical about the consistency between $\langle N_{ret} \rangle$ and $DF \cdot \langle N_{mod} \rangle$.

### 7.4 – Concentration Variability

Rather than draw a standard deviation on each point, in Figs. 10a-d we draw a vertical line (length equal to two standard deviations) centered on the value associated with the median standard deviation. The result for these four medians is representative of what we discussed in Sect. 7.1 and is consistent with our overall finding: variability on $\langle N_{mod} \rangle$ is larger than on $\langle N_{obs} \rangle$, variability on $\langle N_{modw} \rangle$ is intermediate between the observation-based and the unweighted-model values, and variation on $\langle N_{ret} \rangle$ is largest.

### 7.5 – Spectral Dispersion

Here we use the parcel model to evaluate spectral dispersions ($\sigma_D / \langle D \rangle$; Sect. 1). As we discussed in Sect. 6, a parcel model initialized with a distribution of updrafts produces multiple values of $N$ and thus multiple values of $\langle D \rangle$ at a specified height above cloud base. Also, a comparison of these two sets reveals larger $N$ corresponding with smaller $\langle D \rangle$, and vice versa. In this section we derive a spectrum for a cloudy mixture formed by combining the multiple spectra predicted by the parcel model. Implicit in our analysis are two hypotheses: 1)
PDF(w_j) Aw describes the fractional contributions of spectra that enter into the mixture (Sect. 6), and 2) mixing within the cloud produces a homogeneous mixture. In a subsequent step we evaluate σ_D, ⟨D⟩, and the dispersion corresponding to the mixture spectrum. Our hypotheses are linked to two processes: 1) differential cloud base activation (Stevens et al., 1996), and 2) internal mixing, without entrainment (Hudson and Svensson, 1995).

Values of σ_D and ⟨D⟩, derived for the mixture spectra, were analyzed with the values of σ_mod and ⟨N_mod⟩ described previously (Sect. 6). The following theoretical relationship, developed by Cooper (1989), relates these statistics

\[
\frac{\sigma_D}{\langle D \rangle} = \frac{1}{3} \frac{\sigma_{mod}}{\langle N_{mod} \rangle}
\]  

(7)

Fig. 12 is a scatterplot of the spectral dispersions and concentration dispersions derived for the mixture spectra. The slope of the best-fit line is nearly equal to one third. Hence, we conclude that Cooper’s theory and our modeling are consistent. Because Cooper developed his theory with the assumption that parcels experience closed-parcel adiabatic ascent prior to internal mixing, and this is also implicit in our calculations, the consistency is expected. The consistency evident in Fig. 12 is important because it demonstrates that σ_mod, ⟨N_mod⟩ and Eqn. 7 can be used to derive a spectral dispersion for a mixture whose endmembers are defined by adiabatic trajectories. We also note that σ_mod and ⟨N_mod⟩ can be derived with knowledge of PDF(w) and a CCN spectrum (Sect. 6).

Spectral dispersions derived from in-cloud measurements, from our model of differential cloud base activation and internal mixing, and from the retrievals, are summarized in Tab. 6. Presented in the first row are averages and standard deviation for all 44 analysis intervals. Values derived from modeling are presented in the middle-two columns. Left of these are
dispersions derived using the CDP and M-F100 measurements. The second column has the result for spectra that were corrected for instrument broadening. For this we applied the technique developed by Politovich (1993). We evaluated a correction factor for each of the 44 analysis intervals and adjusted the measured values of $\sigma_D$ by subtracting the correction factor in quadrature; in subsequent steps the corrected dispersions were derived and averaged. It is apparent that the correction reduces the measured dispersion by about 25%. Even with the correction (second column), dispersions based on the observations exceed the model (third and fourth columns). Since the clouds were affected by processes other than differential cloud base activation and internal mixing, this exceedance is expected. The fifth column has the result based on Eqn. 7 with values of $\sigma_{obs}$ substituted for $\sigma_{mod}$, and $\langle N_{obs} \rangle$ substituted for $\langle N_{mod} \rangle$. Here there is a large difference, relative to the value shown in the second column, but we do not view this as an inconsistency. Rather, the smaller dispersion derived using $\sigma_{obs}$ and $\langle N_{obs} \rangle$ is consistent, in a qualitative sense, with internal mixing having decreased the concentration variability resulting from differential cloud base activation. The dispersion presented in the sixth column was derived using Eqn. 7 with values of $\sigma_{ret}$ substituted for $\sigma_{mod}$ and with $\langle N_{ret} \rangle$ substituted for $\langle N_{mod} \rangle$. This relatively large dispersion seems consistent with the view that the lidar sensed parcels prior to mixing that occurred within the body of the cloud.

Results presented in Tab. 6 suggest that two model-derived statistics ($\sigma_{mod}$ and $\langle N_{mod} \rangle$), and Eqn. 7, can be used to generate dispersions which are reasonably consistent with corrected dispersions derived from CDP or M-F100 measurements. We are tentative with this conclusion because the correlation of modeled and observed dispersions lacks significance ($r^2 < 0.01$, # = 44, $p = 0.34$). It may be that the horizontal displacement between the in-cloud and
below-cloud segments (~ 100 km) was a significant contributor to the absence of a correlation. This is supported by the fact that the modeled dispersions are more strongly related to 
\[ (1/3) \cdot \sigma_{ret} / \langle N_{ret} \rangle \]
but even here the relationship is not significant at the \( p < 0.05 \) level \( (r^2 = 0.03, \# = 44, p = 0.15) \). We expected better correspondence between model and retrieval because of the simultaneity of the measurements that go into that comparison. This was investigated by correlating a set selected from analysis intervals with large decoupling factors \( (DF > 0.7) \). Compared to intervals with smaller decoupling factors (Sect. 7.3), we expect the selected set to have thermodynamic and CCN properties, at cloud base, more consistent with the below-cloud measurements. In spite of the selection, the subset was also uncorrelated \( (r^2 < 0.01, \# = 18, p = 0.48) \). However, we did find that the average of the ratio of the modeled dispersion divided by \( (1/3) \cdot \sigma_{ret} / \langle N_{ret} \rangle \), for this subset, is physically consistent in the sense that the average is less than one \( (\bar{x} = 0.6 \pm 0.2, \# = 18) \).

In their correlation of modeled and observed spectral standard deviations \( (\sigma_D) \), with the former derived using a model of differential cloud base activation and internal mixing, Hudson et al. (2012) obtained a significant correlation \( (p \leq 0.05) \) for samples with LWC values between 0.86 and 1.61 of adiabatic (their Figure 7b). In addition, Fig. 7b in Hudson et al. (2012) reveals a ratio of the modeled \( \sigma_D \) divided by the observed \( \sigma_D \) about a factor 2 smaller the ratio \( \bar{x} = 0.6 \) we reported in the previous paragraph. Part of this discrepancy is because we formulated our ratio using modeled and observed dispersions while Hudson et al. (2012) formulated theirs using standard deviations \( (\sigma_D) \). Also, the ratios differ because Hudson et al. (2012) did not correct for instrumental broadening and because their measurements were made in maritime cumulus. Cloud type and setting both affect the ratio because updraft tends to be larger in cumulus, while in a
maritime setting, there are generally fewer active CCN (less competition for vapor) at any
prescribed value of $SS$. Both factors increase $SS_{eff}$. If this increase takes the $SS_{eff}$ past a
specified threshold, we expect the modeled concentrations to become less sensitive to updraft,
and the mixture spectrum to be narrower (Warner, 1969; Hudson and Nobel, 2014). This
inference was verified by selecting dispersions from analysis intervals with $SS_{eff} > 0.1 \%$ (Tab.
3); results are shown in the second row of Tab. 6 where it is apparent that the two modeled
dispersions (middle two columns) decreased. A decrease is also evident in the first two columns,
but again the variability is too large to conclude that there is a correlation between modeled and
observed dispersions.

8 - Conclusions

We have presented a statistical analysis of condensational physics occurring within
stratocumulus clouds formed over the southeastern Pacific Ocean. Most of our analysis intervals
consisted of below-cloud and in-cloud flight segments separated by approximately 100 km.
Droplet concentrations retrieved using lidar, and assessments of cloud geometric thickness from
lidar and radar, are the novel aspects of our analysis.

We used a common feature of cloud-topped boundary layers in our analysis (i.e., the $z_{cb}$
- LCL difference, Sect. 5). A decoupling factor, $DF$, was derived and related to the ratio of two
droplet concentrations (Eqn. 6). We used the $DF$ to scale modeled concentrations and obtained
reasonable consistency with observed concentrations. We also documented that the $r^2$ is larger for
the $\langle N_{obs} \rangle$ versus $DF \cdot \langle N_{mod} \rangle$ correlation, compared to the $\langle N_{ret} \rangle$ versus $DF \cdot \langle N_{mod} \rangle$
correlation, and this in spite of the simultaneity of the measurements that entered into the latter
comparison. Error propagating into $\langle N_{ret} \rangle$ was analyzed and shown to range from 5 to 13 \%.
Because experimental error in an averaged droplet concentration measurement can be a factor of 2.3 larger (Snider et al., 2003), we conclude that sources of error, other than those evaluated here, may have affected the retrieved concentrations. Our characterization of LWP, during the C-130 climbs, and our finding that some of these deviated by more than 50% from the adiabatic prediction may be an indication of the additional error incurred in the retrieval. The fact that a change in cloud base, during the climbs, can alias as a departure of the measured LWP from the adiabatic prediction makes dealing with this potential source of error problematic. Measurements from an upward-viewing radiometer operated on the C-130 during VOCALS are available (Ziudema et al., 2012) and may help to minimize the suspected error in the retrieved concentrations.

Considerable headway was made toward understanding the role of differential cloud base activation, and internal mixing, in droplet spectral broadening. Because internal mixing of parcels is assumed, the relevance of our findings to actual clouds is contingent on the degree to which this does occur. Evidence for this is seen in our finding that the quantity $(1/3) \cdot \sigma_{obs}/\langle N_{obs} \rangle$ is substantially smaller than either the dispersion from corrected droplet spectra or the dispersion from modeling (Tab. 6). This has relevance to prior investigations where concentration variability, derived using in-cloud observations, was smaller than that derived with a model of the differential cloud base activation and internal mixing processes (Snider and Brenguier, 2000; Romakkanemi et al., 2009). The present findings suggest that $(1/3) \cdot \sigma_{obs}/\langle N_{obs} \rangle$ may range between an upper limit, which occurs when parcels activating at different updraft speeds are not mixed, to zero in the limit of complete internal mixing.

In addition, we showed that the model-derived dispersion is, on average, 70% of the observation-based dispersion corrected for instrumental broadening. Although this does not
implicate differential cloud base activation and internal mixing as dominant for spectral
broadening, it does suggest that they play an important role. Other relevant processes involve the
entrainment of above-cloud air into the boundary layer (Devenish et al. (2012) and references
therein), internal mixing of droplet-specific trajectories (Cooper, 1989; Politovich, 1993; Cooper
et al., 2013), internal mixing of parcels that reach their LCL’s at different altitudes (Wang et al.,
2009b), adiabatic recirculation of cloudy parcels (Korolev, 1995), and CCN spectral amplitude
and shape (Hudson and Yum, 1997). Since the dispersion is an important factor in predictions of
drizzle formation (Beheng, 1994; Austin et al., 1995; Liu and Daum, 2004) and in predictions of
cloud albedo (Liu and Daum, 2002), and because there is an ongoing debate about how to
parameterize the dispersion in cloud models (Liu and Daum, 2002; Pawlowska et al., 2006;
Hudson and Yum, 1997), we expect that our findings will contribute to the discussion and to
further advancement of stratocumulus microphysics.
Appendix A – Aerosol Particle Hygroscopicity

Information about aerosol hygroscopicity, used in the parcel model, comes from dried-particle and haze-particle spectra recorded during the below-cloud segments. In what follows, we analyze airborne measurements of the dried and humidified spectra using the dual-OPC technique developed by Snider and Petters (2008). At the end of this section we derive the hygroscopicity value we used to model the Köhler curves of particles with dry diameter > 0.11 µm.

For VOCALS, we have improved the dual-OPC technique in two ways. First, laboratory determinations of the OPCs’ (PCASP and F300) response to mobility-classified particles were conducted prior to and after VOCALS. Second, we accounted for a difference between the OPCs’ threshold-diameter relationship derived during lab calibration and that applied during data acquisition on board the C-130. These improvements are documented in Appendix B where we also note that the data archive maintained by NCAR does not account for either correction. The calibrated threshold-diameter relationships (PCASP and F300), assuming refractive index $n = 1.59$ particles, and operation on board the C-130, are provided in Tab. A1. A sizing relationship is also provided for the F300’s assessment of the haze particles; for this we applied the refractive index $n = 1.33$ (Snider and Petters, 2008). For wet diameters greater than 2.5 µm, a few adjacent F300 channels were combined to account for ambiguity due the non-monotonic relationship between particle size and forward scattering intensity.

Fig. A1a presents the PCASP and F300 spectra from the example above-cloud segment. Here we applied the threshold-diameter relationships for $n = 1.59$ particles. The blue and red vertical error bars, most visible on the spectra at $D > 1$ µm, denote the Poissonian sampling uncertainty. Given this uncertainty, it is evident that an inter-probe comparison is not possible
for diameters larger than about 1 μm. Also, for particles smaller than 0.4 μm, comparison is not possible because of the non-physical roll off of the F300 spectrum. Also apparent are power law fits of the spectra (dotted red and blue lines). Following Snider and Petters (2008), these fit lines were used to derive a hygroscopic growth factor (\(GF\)). Provided we have set the sample area of the F300 correctly at \(A_F = 0.10 \text{ mm}^2\) (Appendix B), we expect this \(GF\) to be close to unity in the dry above-cloud air. This is indeed the case for the example presented in Fig. A1a \((RH = 2\%)\). For the 44 above-cloud segments the average \(GF\) is 0.99±0.10.

Fig. A1b presents PCASP and F300 spectra from the example below-cloud segment. As in our analysis of the above-cloud segments, we assume that the PCASP particles are dried and that their refractive index is \(n = 1.59\) (Strapp et al., 1992). Also, consistent with the prior discussion, the haze particles measured by the F300 are assigned a refractive index \(n = 1.33\) and a few channels at \(D > 2.5 \mu m\) are combined to account for ambiguity due the non-monotonic relationship between particle size and forward scattering intensity. The derived \(GF\), written on Fig. A1b, demonstrates that the ambient particles are approximately 40 % larger by virtue of their hygroscopic growth at \(RH = 58\%\).

The 44 determinations of the below-cloud \(GF\) are summarized in Fig. A2a. As in Snider and Petters (2008), these assessments are bounded by predictions which in the upper-limit indicate that the dried particles are equivalent to particles composed of sodium sulfate and in the lower-limit indicate an equivalence to a 60:40 mixture (by mass) of sodium sulfate and a non-hygroscopic component. Consistency with our prior estimates of the hygroscopicity of marine particles (Snider and Petters, 2008) is encouraging but does not preclude the possibility of bias. Possible sources of bias are the OPC calibrations (Appendix B), the OPC measurements and our refractive index assumption. For one of the data values we illustrate the \(GF\) uncertainty
estimated by Snider and Petters (2008). Given the overall consistency of our result with Snider and Petters (2008), our uncertainty estimate may be too conservative.

In Fig. A2b we present the measurements again, but here with a $GF$ prediction based on a value of $\kappa$ pure sodium sulfate (Kreidenweis et al., 2008; $\kappa = 0.74$). A comparison of the solid curves (Figs. A2a-A2b) reveals a $GF$ disagreement for the same aerosol composition (pure Na$_2$SO$_4$). This disagreement can be traced to differences between the water activity parameterizations used to construct the two curves. We note that the difference is quite large at a relative humidity representative of the below-cloud segments, but decreases with increasing $RH$ to a minimal difference at $RH = 90\%$. Consistent with results shown in Fig. A2b, the value $\kappa = 0.74$ was accepted as the project average and was used in the parcel model to describe the Köhler curves of particles with dry diameter > 0.11 µm. These are the particles sized and counted by the PCASP (Tab. A1).
This appendix summarizes our calibrations of two optical particle counters (OPCs). This work was conducted in 2008 and 2011 in our laboratory at the University of Wyoming (UWYO). The instruments are a model 300 Forward Scattering Spectrometer Probe (F300; SN = 25665-0991-05) and a Passive Cavity Axially Scattering Probe (PCASP; SN = 23738-0491-08). Both have the SPP electronics package, developed by Droplet Measurement Technologies (DMT; Boulder, CO), both are owned by the National Center for Atmospheric Research (NCAR), and both are installed in an external pod during airborne operations. During two intervals, in 2008 and 2011, these OPCs were installed on the NCAR C-130 aircraft for the VOCALS (October and November, 2008) and ICET (June and July, 2011) campaigns. The F300 and PCASP were fabricated by Particle Measuring Systems (PMS; Boulder, CO), a predecessor of DMT; the latter company services both instruments.

We describe test particle generation (Sect. B1), data recording (Sect. B2), derived sample area and the channel-diameter relationship we used to analyze C-130 measurements made with the F300 (Sect. B3.1 - B3.1c), and derived aerosol flow and the channel-diameter relationships we used to analyze C-130 measurements made with the PCASP (Sect. B4.1b - B4.1c). The PCASP inlet system is described in Sect. B4.1a.

**B1 – Methods**

Measurements were made in the Department of Atmospheric Science at the University of Wyoming. The aerosol generation system and the aerosol detection instrumentation are shown in Fig. B1. OPC testing was conducted using particles which were size-selected based on their electrical mobility. Test aerosol preparation started with pneumatic atomization of a hydrosol containing polystyrene latex (PSL) spheres. The resulting dispersion was dried, charge
neutralized, size classified in a TSI DMA3081 electrostatic classifier (TSI, 2000) and diluted. In addition to spectra measurements from the OPCs, spectra were measured with a Scanning Mobility Particle Sizer (SMPS) and size-integrated concentrations were measured with a condensation particle counter (CPC). Cai et al. (2013) provide descriptions of the SMPS and CPC used in this testing.

**B2 - Data Acquisition**

The particle count histograms produced by the F300 were recorded using the Particle Analysis and Collection Software (PACS, DMT Inc.); a histogram was recorded every second (1 Hz sampling). Spectra were also obtained using the SMPS (Fig. B1). These were recorded using the Aerosol Instrument Manager software (TSI Inc.) as 300 s average (Cai et al., 2013). In addition, a Labview Virtual Instrument (National Instruments, Inc.) recorded the size-integrated concentrations (CPC), aerosol flowrates (TSI 4010; Fig. B1), the PCASP aerosol flowrates and the PCASP count histograms. The Labview-recorded signals were sampled at 1 Hz.

**B3.1 - F300**

Fig. B1 shows that a convergent tube - 3 mm to 1 mm inner diameter - was used to accelerate particles across the F300’s laser. The position of the tube was adjusted so that the particle count rate, reported by the F300, was a maximum. The particle speed exiting this tube was assumed to be the air speed at the tube’s exit (Fig. B1). Because a range of dilution air flow rates were used in the lab tests (Fig. B1), there were a range of particle speeds; values were between 2 and 25 m/s. Unfortunately the tube’s exit crossection (0.8 mm$^2$) is larger than the F300’s sample area (Sect. B3.1a), so were unable to direct particles exclusively into the sample area.
An outstanding problem with the F300 is the difficulty of determining the portion of its laser beam that produces an unambiguous scattering signal. We note that the F300’s laser beam is oriented perpendicular to the C-130s line of flight and that three dimensions are defined relative to the beam. One of these is along the beam axis (the longitudinal dimension), and the other two are perpendicular (the transverse dimensions). Distances along the three dimensions characterize the F300 sample volume. Also, we note that the probe’s sample volume is defined operationally: particles that produce an in-focus scattering signal pass through the sample volume and those that produce an out-of-focus scattering signal (or negligible scattering signal) do not pass through the sample volume. The two relevant possibilities (in-focus and out-of-focus) are distinguished by the probe’s microprocessor. The basis for the distinction is the microprocessor’s analysis of time-dependent scattering signals reported by a partially masked and unmasked photodetector (Baumgardner et al., 1992).

During airborne operation, one the sample volume’s transverse dimensions is set by the C-130’s true air speed and the data averaging time interval. The latter, in most applications, is 1 s (1 Hz sampling). The sample volume’s two other dimensions are the optical depth-of-field, measured longitudinally, and the second transverse dimension, commonly known as the laser beam height. The product of the depth-of-field and the beam height define the probe’s sample area \( A_F \). Baumgardner et al. (1992) evaluated \( A_F \) by correlating the F300 particle count with particle concentration values reported by a model 100 Forward Scattering Spectrometer Probe. More recently, we determined the \( A_F \) by correlating measurements of F300 count and PCASP concentration (Snider and Petters, 2008).
We applied the technique Snider and Petters (2008) and determined project-averaged $A_F$ values. These are 0.10 mm$^2$ and 0.07 mm$^2$ for VOCALS and ICET, respectively. We note that these determinations of $A_F$ are about a factor of two larger than previously published estimates (Baumgardner et al., 1992; Snider and Petters, 2008). This shift resulted because the previous estimates were based on the manufacturer’s calibration of the threshold-diameter relationship, for the PCASP, and on the manufacturer’s calibration of the PCASP’s aerosol flow system. Both here (Sect. B4.1b and B4.1c), and in Cai et al. (2013), we document significant differences between our calibrations of these probe characteristics and the manufacturer’s. The values of $A_F$ archived in the Network Common Data Format (NetCDF) files, released by NCAR, are about 50% larger than the values $A_F = 0.10$ mm$^2$ (VOCALS) and $A_F = 0.07$ mm$^2$ (ICET) we are recommending here.

**B3.1b - F300 Laboratory Test Data**

F300-derived test particle spectra are presented in the supplementary material (LINK-F300). This figure is a composite of 59 tests. Results are arranged chronologically from May 2009 to August 2011; tests with PSL diameters equal to 343, 491 and 707 nm are reported. Included, for each test, are spectra (300 s average), from the SMPS and F300 (left panel), and the count histogram from the F300 (right panel, also a 300 s average). The vertical dashed line (left panel) is the diameter of the test particles. This diameter is set by the PSL manufacturer’s specification (Duke Scientific Corporation), and by the fact that we select the test particle diameter, at the nominal PSL size, using an electrostatic classifier (Fig. B1).

As summarized in Tab. B1, good agreement was obtained in a comparison of the PSL diameter ($D_{PSL}$) and the mode diameter reported by the SMPS ($D_{SMPS}$). The average relative difference for that comparison is 0.01 or 1 %. Also in Tab. B1, we present a statistical summary.
of the $D_{PSL}$ vs. $D_{F,300}$ and the $D_{PSL}$ vs. $D_{PCASP}$ comparisons. Those results are discussed in Sect. B3.1c and B4.1c, respectively.

From our analysis of the F300 spectra (LINK-F300), we reached two additional conclusions. The first is related to particle charge state within the aerosol generation system. At point “A” (Fig. B1) most of the test particles are singly-charged and most have a diameter equal to the prescribed PSL diameter. When transiting towards the SMPS, the particles pass through a neutralizer, where a Boltzmann charge state is reestablished (TSI, 2000). Subsequent to the neutralizer, and prior to entering the cylinder of the SMPS, at point “B”, both +1 particles and multiply-charged particles (+2, etc.) are present. With knowledge of the PSL particle diameter we calculated the mobility-equivalent diameter of the multiply-charged particles (Snider et al., 2010). Those diameters are indicated with downward arrows in the left-panels of LINK-F300.

In Fig. B2 we present a specific example of the situation described in the previous paragraph. Here the test particles are 491 nm PSL. We see that the F300 and SMPS both respond at the nominal PSL diameter. We also see that particles at 300 nm are detected by the SMPS, but not by the F300. This example makes it clear that that the particles detected by the SMPS, at the diameters indicated by the downward arrows, actually have a diameter equal to $D_{PSL}$, but because they are multiply charged, they are sized at a smaller mobility-equivalent diameter. Such ambiguity is a consequence of the SMPS’s discrimination of particles based on their electrical mobility, and the fact that electrical mobility depends on both a particle’s size and its charge state.

Our second finding relates to a user-selectable option for F300 measurements acquired by the Particle Analysis and Collection Software (PACS) (Sect. B2). When setting up the PACS the user can select either “yes” or “no” for the option Reject-Based-on-Depth-of-Field. In our data
set we have 37 tests with the Reject-Based-on-Depth-of-Field option set to “yes” (in-focus
detections only) and 22 with the option set to “no” (both in-focus and out-of-focus detections).
If “yes” is selected, then only the in-focus detections are registered into the histogram produced
by PACS. If “no” is selected, then all detections (in-focus and out-of-focus) are registered. An
example spectrum, acquired with the option set to “no”, is shown in Fig. B3. It is evident that
most of the F300 detections were classified in channel 0. By comparing Fig. B3 to Fig. B2,
where “yes” is selected, and the test particle size is the same (491 nm), we infer that the in-focus
detections correspond to the minor F300 mode at channel 5 in Fig. B3.

**B3.1c - F300 Sizing Tests**

Spectra corresponding to the “no” tests, and the “yes” tests, and for all test particle sizes
($D_{PSL} = 343, 491$ and 707 nm), were analyzed. Consistent particle sizing results were obtained
for testing conducted in 2008 and in 2011. The supplementary material has 14 tests with 343 nm
particles, 30 tests with 491 nm particles and 15 tests with 707 nm particles (LINK-F300).
Without exception, a histogram mode can be observed in the zeroth channel (343 nm particles),
in the fifth channel (491 nm particles), and in the eighth channel (707 nm particles).

Fig. B4 summarizes the F300 sizing tests we performed and those conducted by DMT.
The instrument has two gain stages and results are split between calibrations for small particles
(high-gain, Fig. B4a) and large particles (low-gain, Fig. B4b). Calibration data points are shown
as triangles with gray and blue indicating calibrations conducted at UWYO and DMT,
respectively. The triangles are plotted at the midpoint of the channel with the maximum
histogram value.

Assignment of an array of thresholds to an array of diameters is contingent on properties
of the OPC (laser illumination, scattering geometry and photodetector signal amplification), and
particle-dependent properties (index of refraction and shape). That assignment is provided by the instrument manufacturer and is referred to as the manufacturer’s calibration. The latter is shown in Figs. B4a-B4b as a dashed black line connecting diamonds plotted at each of the 30 threshold-diameter pairs. Although there are some outliers, we document reasonable agreement between the manufacturer’s calibration and the laboratory testing (i.e., the actual measurements made both in Laramie and at DMT). A statistical comparison of the PSL diameter \( D_{PSL} \) and the F300’s channel midpoint diameter, based on the manufacturer’s calibration, is shown in Tab. B1. It is apparent that the average relative difference is 0.09 or 9%. Compared to the result seen in the second row of Tab. B1 (0.01 or 1%), the average relative difference is larger. This reflects both the scatter of measurements about manufacturer’s calibration (Figs. B4a and B4b) and the fact that the sizing resolution of the F300 is coarser than that of the SMPS (Cai et al., 2013).

Given the reasonable consistency between laboratory testing and the manufacturer’s calibration, we applied the manufacturer’s threshold-diameter calibration to the C-130 measurements made in VOCALS and ICET. However, an adjustment is needed to fully incorporate our laboratory finding with the C-130 measurements.

The analysis discussed in the previous paragraph is based on measurements made with the F300 initialized with the manufacturer’s threshold-diameter table. When the instrument is operated on the C-130 a non-conventional threshold-diameter table is used (private communication David Rogers, April 16, 2009). The C-130 threshold-diameter table is presented in Tab. A1. In Figs. B4a-B4b, the vertical red-dashed lines illustrate the C-130 thresholds. It is apparent, particularly in Fig. B4a, that we evaluated the C-130 calibration diameters at the intersections of the C-130 thresholds (vertical red-dashed line) and the manufacturer’s threshold-diameter calibration.
A set of calibration diameters, somewhat different from that in Tab. A1, was archived in the NetCDF files released by NCAR. For example, the diameters in the NetCDF archive of the VOCALS campaign are 4 to 50% larger than our recommendation in Tab. A1. Users of the VOCALS and ICET data sets are encouraged to use the C-130 threshold-diameter table provided in Tab. A1. These are also the basis for our analysis of the VOCALS C-130 F300 measurements.

**B4.1 - PCASP**

**B4.1a - PCASP Heating of the Aerosol Sample**

The PCASP’s airflow system is designed to direct an aerosol stream across the probe’s Helium-Neon laser ($\lambda = 0.633$ μm). Particle loss is minimized by directing the stream along a straight path from the sample inlet to the laser (Fig. B5). The aerosol stream first encounters the PCASP at the diffuser, where it is decelerated from the C-130’s true airspeed (~110 m/s) to ~11 m/s (Particle Measuring Systems, 2002). The aerosol stream then passes through a narrow tube (inner diameter = 0.5 mm), where it is combined with sheath air; the combined flow then crosses the laser. The volumetric rate of the sheath stream is set to be 15 times the aerosol flowrate (Particle Measuring Systems, 2002). Because of a nozzle restriction at the point where the flows are combined, the combined stream crosses the laser at approximately 45 m/s (Particle Measuring Systems, 2002). The combined stream exits the sample cavity to a pump, is passed through a tube filled with granular desiccant, then through a filter, and is split. One of the streams is passed through the sheath flow valve, is monitored in a mass flow meter, and recirculated. The other stream is passed through the sample flow valve, is monitored in a mass flow meter, and is dumped. Because of mass continuity, the mass flowrate monitored by the aerosol flow meter, is equivalent to the aerosol mass flowrate passing through the sample cavity.
The devices used for the sheath and aerosol flow measurement are the Model AWM3300V and AWM3100V mass flow meters (Honeywell).

In the previous paragraph we mentioned the tube that carries the aerosol stream from the diffuser to the sample cavity. This tube is evident in Fig. B5 and will be referred to as the “needle.” The PCASP inlet is equipped with three deice heaters. These are automatically activated when the C-130 leaves the runway. The heaters are located near the tip of the diffuser (35 Watt), at the back of the diffuser (100 Watt), and in close proximity to the front end of the needle (10 Watt). Strapp et al. (1992) demonstrated that compressional warming, combined with heating due to the deice heaters, can have a substantial effect on the size of wet aerosol particles sampled by the PCASP. They estimated that particles reside sufficiently long within the decelerating stream, and within the probe, to lose most of their chemically-bound water. Strapp et al. (1992) estimated the interaction time to be 0.2 s. Snider and Petters (2008) used a model similar to that employed by Strapp et al. (1992) and showed that particles starting at a wet diameter 0.84 µm, corresponding to a haze particle equilibrated at $RH = 96\%$, evaporate to 0.48 µm. In Strapp et al. (1992) and in Snider and Petters (2008), model relative humidities ≤ 40 % were assumed. Measurements are needed to validate this probe- $RH$ assumption.

B4.1b – PCASP Aerosol Flow Meter Calibration

The PCASP derives particle concentration as the ratio of the particle count rate (number of particles per second) and aerosol flowrate (actual cubic centimeter per second). The latter is derived in two steps. First, the signal from the PCASP’s aerosol flow sensor, represented either as an analog signal (millivolt, $mV$; VOCALS), or as an integer (cnt; ICET), is used to derive the aerosol flowrate (standard cubic centimeter per second, sccps). The project-specific calibrations are
sccps = -0.0165 + (7.9354e-05) · mV + 1.1453e-07 · mV²  
(VOCALS)  (B1)

and

cscps = 7.51885 + (-8.46821e-3) · cnt + 2.30130e-6 · cnt²  
(ICET)  (B2)

In the second step, the standard flowrate value is converted to the ambient flowrate. This is done by evoking the ideal gas law with measurements of ambient pressure and temperature.

We evaluated the flowrate calibrations (Eqns. B1-2) by correlating flow measurements (converted to standard pressure and temperature) with the signal output by the PCASP’s aerosol flow meter. The integer (cnt) and analog (mV) signals are relatable via a 12 bit analog-to-digital conversion.

B4.1c - PCASP Sizing Calibration

PCASP’s response to a known particle size was evaluated using PSL test particles ($D_{PSL} = 125, 152, 199, 491$ and 707 nm); results from 47 tests are shown in the supplementary material (LINK-PCASP). Evident in the left panels is a vertical dashed line, plotted at the diameter of the PSL test particle, and arrows indicating the mobility-equivalent diameter of the multiply-charged test particles. The latter are described in Sect. B3.1b.

Fig. B6 summarizes the PCASP sizing calibrations. The probe has three gain stages, and therefore the results are split to show calibrations of the high-gain (Fig. B6a), mid-gain (Fig. B6b), and low-gain stages (Fig. B6c). Calibration data points are indicated as filled circles; these are plotted at the midpoint of the channel with the maximum histogram value. For all gain stages, and especially for the high-gain stage (Fig. B6a), we find that the manufacturer’s calibration (dashed line connecting diamonds) underestimates particle diameter. Because of this, we have provided an adjusted threshold-diameter calibration (solid line). A statistical comparison of laboratory-determinations of particle size, based on the adjusted calibration (Cai
et al., 2013), is shown in Tab. B1. It is apparent that the average relative difference is 0.03 or 3%

The analysis summarized in the fourth row of Tab. B1 is based on measurements made with the PCASP initialized with the adjusted threshold-diameter table. When the instrument is operated on the C-130 a non-conventional threshold table is used (private communication Allen Schanot, June 12, 2009). The C-130 thresholds are presented in Figs. B6a-B6c as vertical red-dashed lines. It is apparent, particularly in Fig. B6a, that we evaluated the calibration diameters at the intersections of the C-130 thresholds and the adjusted threshold-diameter table. At their greatest absolute relative departure, these calibrated diameters are 36% smaller than the values archived by NCAR for VOCALS, and 12% larger than the values released for ICET. More typical relative departures are -5% (VOCALS) and +5% (ICET). Users of the VOCALS and ICET data sets are encouraged to use the threshold-diameter table provided in Tab. A1. These are also the basis for our analysis of the VOCALS C-130 PCASP measurements.
Fig. 1 – Sampling segments (above-cloud, in-cloud, and below-cloud) for the example analysis interval (RF04). a) Aircraft track; b) Aircraft altitude.
CDP (RF12, RF13, RF14)  
\( <N_{\text{obs}} > = 168. \, \sigma_{\text{obs}} = 78. \, f_a = 1.00 \)

CDP (RF12, RF13, RF14) - Corrected  
\( <N_{\text{obs}} > = 208. \, \sigma_{\text{obs}} = 107. \, f_a = 1.00 \)

U-F100 (RF12, RF13, RF14)  
\( <N_{\text{obs}} > = 140. \, \sigma_{\text{obs}} = 62. \, f_a = 0.80 \)

Fig. 2 – Droplet concentration PDFs from RF12, RF13 and RF14. Two CDP ensembles (black = without the L12 correction and red = with the L12 correction), and one U-F100 ensemble (gray), are presented. Properties of the data ensembles (average, standard deviation, and fraction accepted) are presented in the legend.
CDP (RF12, RF13, RF14)
\[ <\sigma_g> = 1.4 \quad \sigma = 0.1 \quad fa = 1.00 \]
Overall (See text for explanation)
\[ <\sigma_g> = 1.3 \quad \sigma = 0.2 \quad fa = 0.97 \]
U-F100 (RF12, RF13, RF14)
\[ <\sigma_g> = 1.3 \quad \sigma = 0.1 \quad fa = 0.80 \]

Fig. 3 – Geometric standard deviation PDFs from RF12, RF13 and RF14 (black = CDP and gray = U-F100), and the overall \( \sigma_g \) PDF (orange). The latter is based on measurements from 44 in-cloud segments (11 flights) with the M-F100 substituted for the CDP in RF04. Properties of the data ensembles (average, standard deviation, and fraction accepted) are presented in the legend.
Fig. 4 – a) The example in-cloud vertical velocity sequence, b) example below-cloud vertical velocity sequence, and c) the in-cloud and below-cloud vertical velocity PDFs.
Fig. 5 - a) Below-cloud CCN measurements and fit line of form $n(SS) = C \cdot SS^k$ for the example interval (RF05, 10:28-10:41 UTC). b) The combined spectrum for the example interval. At $SS \leq 0.11\%$, the spectrum was derived using the Köhler model with PCASP measurements and a kappa. At $SS > 0.11\%$, the spectrum is from the fit of measurements made with the Wyoming CCN instrument (Fig. 5a). See Sect. 4.4 for details.
Fig. 6 – Properties retrieved by upward-viewing remote sensors during the example below-cloud sequence (RF05, 10:28-10:41 UTC). a) Radar-derived $z_{ct}$ and lidar-derived $z_{cb}$. b) Upward-viewing radiative temperature (RSTT) and lidar-derived cloud base temperature ($T_{cb}$). c) Lidar-derived droplet concentration ($N_{ret}$). All properties were resampled at 1 Hz for plotting clarity. See text for details.
Fig. 7 – Relationship between distance above cloud base and extinction coefficient for different values of \( N \) and \( \sigma_g \). The gray regions are defined by \( N \) equal to 20 cm\(^{-3}\) and 200 cm\(^{-3}\). The left and right boundaries of these regions are at \( \sigma_g = 1.5 \) and \( \sigma_g = 1.1 \), respectively. The left and right red curves, extending upward from 20 m, are extinction profiles derived with \( N_o = 21.5 \) cm\(^{-3}\) and \( \sigma_{g,o} = 1.3 \), and \( N_o = 212 \) cm\(^{-3}\) and \( \sigma_{g,o} = 1.3 \), respectively. The red curves intersect hypothetical lidar-derived states (red triangles) which can also be matched by \( N = 20 \) cm\(^{-3}\) and \( \sigma_g = 1.2 \) (left) and by \( N = 200 \) cm\(^{-3}\) and \( \sigma_g = 1.2 \) (right).
Fig. 8 – a) Distribution of temperature difference based on cloud base temperature from the lidar $(T_o = T_{cb})$ and from the upward viewing radiometric temperature sensor $(T = RSTT)$. The “Example” is the 10:28 to 10:41 UTC below-cloud segment from RF05. Values from the 44 analysis intervals are represented in the “Overall” distribution. b) Distribution of LWC lapse rate difference (cloud base pressure is set equal to 900 hPa). c) Distribution of the relative error in retrieved $N$ using Eqn. 4a (with the $\sigma_g$ derivative set to zero), Eqn. 4b, and the distributions in Fig. 8b.
Fig. 9 – Concentration PDFs for the example in-cloud segment (RF05, 10:13-10:21 UTC) and the example below-cloud segment (RF05, 10:28-10:41 UTC). a) The observation-based and retrieval-based PDFs. b) The unweighted model and updraft-weighted model PDFs.
Fig. 10 – a) Observed cloud droplet concentration ($\langle N_{obs} \rangle$) versus longitude and best-fit line. The data point with an error bar is the one with the median width. b) As in Fig. 10a, but for the retrieved concentration ($\langle N_{ret} \rangle$). c) As in Fig. 10a, but for the unweighted model concentration ($\langle N_{mod} \rangle$). d) As in Fig. 10a, but for the updraft-weighted model concentration ($\langle N_{mod,w} \rangle$).
Fig. 11 – Black circles are the observed-modeled concentration pairs and gray circles are observed-modeled concentration pairs with the latter adjusted by the decoupling factor $DF$. For the black and gray circles the $Y = a \cdot X$ best-fit coefficients are $a = 0.70 \pm 0.04$ (# = 44) and $a = 1.01 \pm 0.09$ (# = 38), respectively. For both regression lines the values plotting on the ordinate were used to evaluate departures from the fit line and the margin of error associated with the fitted coefficient. The latter were evaluated at the 95 % confidence level (Havilcek and Crain, 1988).
Fig. 12 – On the ordinate are spectral dispersions and on the abscissa are ratios of the standard deviation and average of the concentration PDF. The ordinate and abscissa values were derived from parcel model simulations (unweighted model) with the in-cloud mixing hypothesis. The $Y = a \cdot X$ best-fit coefficient is $a = 0.35 \pm 0.01$ ($\# = 44$) and the dashed line represents the fit equation. Values plotting on the ordinate were used to evaluate departures from the fit line and the margin of error associated with the fitted coefficient. The latter were evaluated at the 95% confidence level (Havilcek and Crain, 1988).
Fig. A1 – a) Above-cloud spectra, PCASP and F300, for the example above-cloud segment (RF05, 10:00-10:10 UTC). Particle refractive index ($n$) is assumed equal to 1.59 in both OPCs.

b) Below-cloud spectra, PCASP and F300, for the example below-cloud segment (RF05, 10:28-10:41 UTC). Particle refractive index is assumed at $n = 1.59$ and $n = 1.33$ in the PCASP and F300, respectively.
Fig. A2 – a) GF measurements versus RH and model predictions based on Snider et al. (2003) with two hygroscopic mass fractions ($\varepsilon = 1$ and $\varepsilon = 0.6$), the Na$_2$SO$_4$ parameterization from Tang and Munkelwitz (1994), and density of the non-hygroscopic fraction equal to 1200 kg/m$^3$. b) GF measurements and model prediction based on Petters and Kreidenweis (2007; their Eqn. 11) with $\kappa = 0.74$ for Na$_2$SO$_4$ (Kreidenweis et al., 2008). The GF uncertainty from Snider and Petters (2008) is also indicated for one of the data points.
Fig. B1 – Schematic diagram of the particle generation and measurement systems
Fig. B2 – Laboratory spectra selected from the set of 59. The test particles are mobility-selected PSL particles at $D_{PSL} = 491$ nm (vertical dashed line in left panel). Downward arrows indicate the mobility-equivalent size of multiply-charged 491 nm PSL particles. The Reject-Based-on-Depth-of-Field option is set to “yes.” See text for details.
Fig. B3 – As in Fig. B2, but with the Reject-Based-on-Depth-of-Field option set to “no.” The test particles are mobility-selected PSL particles at $D_{PSL} = 491$ nm (vertical dashed line in left panel).
Fig. B4 – Summary of F300-based particle sizing tests performed at DMT and UWYO.

Experimental determinations of the channel that PSL spheres classify into (blue and gray triangles) are plotted at the midpoint of the channel with the maximum histogram value. The manufacturer’s threshold-diameter table (dotted black line connecting diamonds) and the C-130 thresholds (vertical red-dashed lines) are also illustrated.
Fig. B5 - The PCASP's sampling system (Particle Measuring Systems, 2002).
Fig. B6 – Summary of PCASP-based particle sizing performed at the University of Wyoming. Illustrated are the manufacturer’s threshold-diameter table (dotted black line connecting diamonds), the C-130 thresholds (vertical red-dashed lines), and experimental determinations of the channels that PSL spheres (0.152 µm, 0.199 µm, and 0.491 µm) classify into (filled circles). The latter are plotted at the midpoint of the channel with the maximum histogram value. See text for details.
Table 1 – Symbols used for cloud droplet concentration and width

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Location of relevant measurements</th>
<th>Equation or section where defined</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N$</td>
<td>Generic cloud droplet concentration</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$&lt;N_{\text{obs}}&gt;$ and $\sigma_{\text{obs}}$</td>
<td>In-cloud observed average concentration and standard deviation</td>
<td>In-cloud segment</td>
<td>Sect. 4.1</td>
</tr>
<tr>
<td>$&lt;N_{\text{ret}}&gt;$ and $\sigma_{\text{ret}}$</td>
<td>Lidar-retrieved average concentration and standard deviation</td>
<td>Below-cloud segment</td>
<td>Sect. 5</td>
</tr>
<tr>
<td>$&lt;N_{\text{mod}}&gt;$ and $\sigma_{\text{mod}}$</td>
<td>Unweighted model average concentration and standard deviation</td>
<td>Below-cloud segment (CCN spectrum and updraft)</td>
<td>Eqns. 5a and 5c</td>
</tr>
<tr>
<td>$&lt;N_{\text{mod,w}}&gt;$ and $\sigma_{\text{mod,w}}$</td>
<td>Updraft-weighted model average concentration and standard deviation</td>
<td>Below-cloud segment (CCN spectrum and updraft)</td>
<td>Eqns. 5b and 5d</td>
</tr>
</tbody>
</table>
Table 2 – Analysis Intervals, averages and standard deviations. 

1 Flight number; 2 Average observed droplet concentration; 3 Standard deviation of observed droplet concentration; 4 Average vertical velocity (below-cloud segments); 5 Standard deviation of vertical velocity (below-cloud segments); 6 Fitted parameter in n(SS) = C·SS^k; 7 Fitted parameter in n(SS) = C·SS^k; 8 Average retrieved droplet concentration; 9 Standard deviation of retrieved droplet concentration; 10 Average modeled droplet concentration (Eqn. 5a); 11 Standard deviation of modeled droplet concentration (Eqn. 5b); 12 Average modeled droplet concentration (Eqn. 5c); 13 Standard deviation of modeled droplet concentration (Eqn. 5d).

<table>
<thead>
<tr>
<th>RF</th>
<th>Flight number</th>
<th>In-cloud Start UTC hhmm</th>
<th>In-cloud End UTC hhmm</th>
<th>In-cloud</th>
<th>&lt;N_\text{obs}&gt;</th>
<th>\sigma_\text{obs}</th>
<th>Below-cloud Start UTC hhmm</th>
<th>Below-cloud End UTC hhmm</th>
<th>&lt;w&gt;</th>
<th>\sigma_w</th>
<th>C</th>
<th>k</th>
<th>&lt;N_\text{ret}&gt;</th>
<th>\sigma_\text{ret}</th>
<th>&lt;N_\text{mod}&gt;</th>
<th>\sigma_\text{mod}</th>
<th>&lt;N_\text{mod, w}&gt;</th>
<th>\sigma_\text{mod, w}</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1937</td>
<td>1946</td>
<td>183</td>
<td>62</td>
<td>1914</td>
<td>1927</td>
<td>0.09</td>
<td>0.43</td>
<td>482</td>
<td>0.42</td>
<td>177</td>
<td>93</td>
<td>249</td>
<td>67</td>
<td>280</td>
<td>23</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>1806</td>
<td>1811</td>
<td>155</td>
<td>36</td>
<td>1747</td>
<td>1757</td>
<td>0.03</td>
<td>0.40</td>
<td>575</td>
<td>0.48</td>
<td>138</td>
<td>90</td>
<td>226</td>
<td>86</td>
<td>283</td>
<td>46</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>1848</td>
<td>1856</td>
<td>158</td>
<td>35</td>
<td>1829</td>
<td>1843</td>
<td>0.03</td>
<td>0.40</td>
<td>920</td>
<td>0.20</td>
<td>168</td>
<td>99</td>
<td>438</td>
<td>196</td>
<td>571</td>
<td>117</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1338</td>
<td>1342</td>
<td>316</td>
<td>29</td>
<td>1346</td>
<td>1357</td>
<td>0.24</td>
<td>0.45</td>
<td>970</td>
<td>0.35</td>
<td>218</td>
<td>115</td>
<td>437</td>
<td>151</td>
<td>527</td>
<td>77</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1415</td>
<td>1423</td>
<td>271</td>
<td>32</td>
<td>1428</td>
<td>1438</td>
<td>0.15</td>
<td>0.40</td>
<td>920</td>
<td>0.20</td>
<td>168</td>
<td>99</td>
<td>438</td>
<td>196</td>
<td>571</td>
<td>117</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1510</td>
<td>1514</td>
<td>203</td>
<td>48</td>
<td>1523</td>
<td>1533</td>
<td>0.12</td>
<td>0.40</td>
<td>230</td>
<td>0.10</td>
<td>111</td>
<td>55</td>
<td>165</td>
<td>51</td>
<td>192</td>
<td>17</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1551</td>
<td>1600</td>
<td>149</td>
<td>30</td>
<td>1605</td>
<td>1608</td>
<td>0.12</td>
<td>0.40</td>
<td>222</td>
<td>0.05</td>
<td>96</td>
<td>51</td>
<td>174</td>
<td>47</td>
<td>201</td>
<td>15</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1812</td>
<td>1825</td>
<td>110</td>
<td>24</td>
<td>1710</td>
<td>1720</td>
<td>0.02</td>
<td>0.40</td>
<td>176</td>
<td>0.08</td>
<td>271</td>
<td>116</td>
<td>342</td>
<td>69</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>0650</td>
<td>0658</td>
<td>167</td>
<td>28</td>
<td>0703</td>
<td>0712</td>
<td>0.13</td>
<td>0.42</td>
<td>350</td>
<td>0.24</td>
<td>118</td>
<td>67</td>
<td>187</td>
<td>82</td>
<td>228</td>
<td>59</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>0731</td>
<td>0740</td>
<td>85</td>
<td>16</td>
<td>0745</td>
<td>0755</td>
<td>0.04</td>
<td>0.29</td>
<td>325</td>
<td>0.47</td>
<td>82</td>
<td>46</td>
<td>129</td>
<td>75</td>
<td>172</td>
<td>56</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>0828</td>
<td>0833</td>
<td>93</td>
<td>18</td>
<td>0839</td>
<td>0849</td>
<td>0.04</td>
<td>0.29</td>
<td>384</td>
<td>0.42</td>
<td>50</td>
<td>38</td>
<td>172</td>
<td>87</td>
<td>222</td>
<td>59</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>0911</td>
<td>0920</td>
<td>81</td>
<td>25</td>
<td>0926</td>
<td>0936</td>
<td>0.02</td>
<td>0.35</td>
<td>346</td>
<td>0.38</td>
<td>75</td>
<td>61</td>
<td>161</td>
<td>64</td>
<td>199</td>
<td>28</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>1011</td>
<td>1012</td>
<td>65</td>
<td>16</td>
<td>1020</td>
<td>1030</td>
<td>0.04</td>
<td>0.38</td>
<td>301</td>
<td>0.47</td>
<td>53</td>
<td>56</td>
<td>136</td>
<td>53</td>
<td>171</td>
<td>23</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>1100</td>
<td>1103</td>
<td>39</td>
<td>9</td>
<td>1109</td>
<td>1118</td>
<td>0.04</td>
<td>0.34</td>
<td>318</td>
<td>0.39</td>
<td>24</td>
<td>16</td>
<td>156</td>
<td>61</td>
<td>183</td>
<td>38</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>1149</td>
<td>1158</td>
<td>79</td>
<td>17</td>
<td>1204</td>
<td>1214</td>
<td>0.01</td>
<td>0.38</td>
<td>254</td>
<td>0.28</td>
<td>145</td>
<td>179</td>
<td>122</td>
<td>69</td>
<td>165</td>
<td>52</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>1233</td>
<td>1243</td>
<td>88</td>
<td>20</td>
<td>1248</td>
<td>1258</td>
<td>0.01</td>
<td>0.36</td>
<td>432</td>
<td>0.38</td>
<td>122</td>
<td>100</td>
<td>198</td>
<td>108</td>
<td>261</td>
<td>79</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0707</td>
<td>0710</td>
<td>231</td>
<td>66</td>
<td>0721</td>
<td>0731</td>
<td>0.16</td>
<td>0.37</td>
<td>468</td>
<td>0.10</td>
<td>256</td>
<td>206</td>
<td>289</td>
<td>131</td>
<td>372</td>
<td>60</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0747</td>
<td>0748</td>
<td>90</td>
<td>10</td>
<td>0803</td>
<td>0813</td>
<td>0.08</td>
<td>0.39</td>
<td>199</td>
<td>0.24</td>
<td>169</td>
<td>167</td>
<td>123</td>
<td>33</td>
<td>142</td>
<td>14</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0846</td>
<td>0855</td>
<td>90</td>
<td>18</td>
<td>0901</td>
<td>0911</td>
<td>0.16</td>
<td>0.43</td>
<td>178</td>
<td>0.09</td>
<td>82</td>
<td>77</td>
<td>137</td>
<td>35</td>
<td>156</td>
<td>14</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0931</td>
<td>0939</td>
<td>94</td>
<td>19</td>
<td>0945</td>
<td>0954</td>
<td>0.02</td>
<td>0.48</td>
<td>205</td>
<td>0.26</td>
<td>92</td>
<td>87</td>
<td>123</td>
<td>39</td>
<td>148</td>
<td>18</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>1013</td>
<td>1021</td>
<td>89</td>
<td>17</td>
<td>1028</td>
<td>1041</td>
<td>0.05</td>
<td>0.55</td>
<td>209</td>
<td>0.48</td>
<td>77</td>
<td>49</td>
<td>104</td>
<td>33</td>
<td>128</td>
<td>22</td>
<td></td>
<td></td>
</tr>
<tr>
<td>----</td>
<td>--------</td>
<td>--------</td>
<td>------</td>
<td>------</td>
<td>--------</td>
<td>--------</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>1113</td>
<td>1117</td>
<td>86</td>
<td>21</td>
<td>1127</td>
<td>1137</td>
<td>0.05</td>
<td>0.47</td>
<td>254</td>
<td>0.61</td>
<td>107</td>
<td>121</td>
<td>110</td>
<td>37</td>
<td>135</td>
<td>26</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>1159</td>
<td>1207</td>
<td>99</td>
<td>14</td>
<td>1212</td>
<td>1223</td>
<td>0.02</td>
<td>0.44</td>
<td>218</td>
<td>0.35</td>
<td>77</td>
<td>59</td>
<td>115</td>
<td>37</td>
<td>143</td>
<td>23</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>1256</td>
<td>1306</td>
<td>96</td>
<td>13</td>
<td>1312</td>
<td>1323</td>
<td>0.04</td>
<td>0.45</td>
<td>193</td>
<td>0.23</td>
<td>99</td>
<td>48</td>
<td>127</td>
<td>29</td>
<td>144</td>
<td>14</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>1343</td>
<td>1353</td>
<td>154</td>
<td>29</td>
<td>1357</td>
<td>1407</td>
<td>0.06</td>
<td>0.46</td>
<td>357</td>
<td>0.27</td>
<td>144</td>
<td>101</td>
<td>210</td>
<td>61</td>
<td>245</td>
<td>27</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>1439</td>
<td>1449</td>
<td>228</td>
<td>24</td>
<td>1454</td>
<td>1500</td>
<td>-0.03</td>
<td>0.35</td>
<td>591</td>
<td>0.17</td>
<td>120</td>
<td>55</td>
<td>290</td>
<td>144</td>
<td>390</td>
<td>78</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>1036</td>
<td>1045</td>
<td>97</td>
<td>21</td>
<td>0934</td>
<td>0938</td>
<td>0.07</td>
<td>0.53</td>
<td>160</td>
<td>0.10</td>
<td>98</td>
<td>50</td>
<td>125</td>
<td>26</td>
<td>139</td>
<td>9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>1229</td>
<td>1238</td>
<td>93</td>
<td>22</td>
<td>1243</td>
<td>1248</td>
<td>-0.16</td>
<td>0.47</td>
<td>325</td>
<td>0.41</td>
<td>91</td>
<td>42</td>
<td>148</td>
<td>40</td>
<td>173</td>
<td>27</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>1313</td>
<td>1318</td>
<td>143</td>
<td>22</td>
<td>1326</td>
<td>1334</td>
<td>-0.02</td>
<td>0.33</td>
<td>355</td>
<td>0.40</td>
<td>130</td>
<td>108</td>
<td>216</td>
<td>93</td>
<td>275</td>
<td>43</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>0813</td>
<td>0822</td>
<td>145</td>
<td>21</td>
<td>0829</td>
<td>0836</td>
<td>0.10</td>
<td>0.47</td>
<td>286</td>
<td>0.01</td>
<td>136</td>
<td>79</td>
<td>161</td>
<td>84</td>
<td>209</td>
<td>59</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>1117</td>
<td>1124</td>
<td>263</td>
<td>39</td>
<td>1013</td>
<td>1017</td>
<td>0.10</td>
<td>0.33</td>
<td>829</td>
<td>0.21</td>
<td>185</td>
<td>94</td>
<td>386</td>
<td>197</td>
<td>523</td>
<td>113</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>1254</td>
<td>1302</td>
<td>171</td>
<td>28</td>
<td>1308</td>
<td>1318</td>
<td>-0.01</td>
<td>0.29</td>
<td>534</td>
<td>0.20</td>
<td>180</td>
<td>94</td>
<td>237</td>
<td>152</td>
<td>340</td>
<td>123</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>1358</td>
<td>1408</td>
<td>179</td>
<td>40</td>
<td>1345</td>
<td>1352</td>
<td>-0.03</td>
<td>0.31</td>
<td>304</td>
<td>0.13</td>
<td>102</td>
<td>55</td>
<td>177</td>
<td>84</td>
<td>217</td>
<td>59</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>1017</td>
<td>1026</td>
<td>72</td>
<td>28</td>
<td>1032</td>
<td>1041</td>
<td>0.07</td>
<td>0.35</td>
<td>301</td>
<td>0.21</td>
<td>103</td>
<td>77</td>
<td>164</td>
<td>68</td>
<td>209</td>
<td>34</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>1100</td>
<td>1109</td>
<td>76</td>
<td>25</td>
<td>1114</td>
<td>1124</td>
<td>0.02</td>
<td>0.38</td>
<td>447</td>
<td>0.30</td>
<td>130</td>
<td>58</td>
<td>207</td>
<td>84</td>
<td>265</td>
<td>49</td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>1325</td>
<td>1335</td>
<td>265</td>
<td>49</td>
<td>1339</td>
<td>1348</td>
<td>0.12</td>
<td>0.45</td>
<td>671</td>
<td>0.10</td>
<td>154</td>
<td>88</td>
<td>402</td>
<td>170</td>
<td>522</td>
<td>99</td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>1501</td>
<td>1510</td>
<td>157</td>
<td>33</td>
<td>1514</td>
<td>1524</td>
<td>0.12</td>
<td>0.29</td>
<td>515</td>
<td>0.34</td>
<td>89</td>
<td>64</td>
<td>209</td>
<td>91</td>
<td>274</td>
<td>60</td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>1551</td>
<td>1600</td>
<td>150</td>
<td>35</td>
<td>1602</td>
<td>1616</td>
<td>0.04</td>
<td>0.18</td>
<td>856</td>
<td>0.33</td>
<td>98</td>
<td>67</td>
<td>182</td>
<td>116</td>
<td>285</td>
<td>117</td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>1425</td>
<td>1428</td>
<td>148</td>
<td>17</td>
<td>1432</td>
<td>1442</td>
<td>0.11</td>
<td>0.26</td>
<td>175</td>
<td>0.34</td>
<td>113</td>
<td>86</td>
<td>97</td>
<td>54</td>
<td>125</td>
<td>41</td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>1521</td>
<td>1522</td>
<td>51</td>
<td>8</td>
<td>1525</td>
<td>1535</td>
<td>0.14</td>
<td>0.29</td>
<td>64</td>
<td>0.39</td>
<td>60</td>
<td>111</td>
<td>40</td>
<td>8</td>
<td>44</td>
<td>5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>1815</td>
<td>1817</td>
<td>16</td>
<td>5</td>
<td>1704</td>
<td>1709</td>
<td>0.06</td>
<td>0.31</td>
<td>86</td>
<td>0.25</td>
<td>46</td>
<td>40</td>
<td>51</td>
<td>16</td>
<td>61</td>
<td>9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>1515</td>
<td>1525</td>
<td>196</td>
<td>49</td>
<td>1529</td>
<td>1534</td>
<td>0.13</td>
<td>0.29</td>
<td>370</td>
<td>0.21</td>
<td>97</td>
<td>40</td>
<td>207</td>
<td>71</td>
<td>248</td>
<td>34</td>
<td></td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>1740</td>
<td>1744</td>
<td>49</td>
<td>8</td>
<td>1652</td>
<td>1706</td>
<td>0.05</td>
<td>0.22</td>
<td>281</td>
<td>0.46</td>
<td>57</td>
<td>68</td>
<td>105</td>
<td>44</td>
<td>136</td>
<td>31</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
## Table 3 – Effective supersaturation for each analysis interval

<table>
<thead>
<tr>
<th>RF</th>
<th>Below-cloud start, UTC hhmm</th>
<th>Below-cloud end, UTC hhmm</th>
<th>SS_{eff}</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1914</td>
<td>1927</td>
<td>0.073</td>
</tr>
<tr>
<td>1</td>
<td>1747</td>
<td>1757</td>
<td>0.073</td>
</tr>
<tr>
<td>1</td>
<td>1829</td>
<td>1843</td>
<td>0.070</td>
</tr>
<tr>
<td>2</td>
<td>1346</td>
<td>1357</td>
<td>0.060</td>
</tr>
<tr>
<td>2</td>
<td>1428</td>
<td>1438</td>
<td>0.046</td>
</tr>
<tr>
<td>2</td>
<td>1523</td>
<td>1533</td>
<td>0.043</td>
</tr>
<tr>
<td>2</td>
<td>1605</td>
<td>1608</td>
<td>0.068</td>
</tr>
<tr>
<td>2</td>
<td>1710</td>
<td>1720</td>
<td>0.051</td>
</tr>
<tr>
<td>2</td>
<td>1902</td>
<td>1911</td>
<td>0.045</td>
</tr>
<tr>
<td>4</td>
<td>0703</td>
<td>0712</td>
<td>0.061</td>
</tr>
<tr>
<td>4</td>
<td>0745</td>
<td>0755</td>
<td>0.047</td>
</tr>
<tr>
<td>4</td>
<td>0839</td>
<td>0849</td>
<td>0.045</td>
</tr>
<tr>
<td>4</td>
<td>0926</td>
<td>0936</td>
<td>0.052</td>
</tr>
<tr>
<td>4</td>
<td>1020</td>
<td>1030</td>
<td>0.045</td>
</tr>
<tr>
<td>4</td>
<td>1109</td>
<td>1118</td>
<td>0.033</td>
</tr>
<tr>
<td>4</td>
<td>1204</td>
<td>1214</td>
<td>0.045</td>
</tr>
<tr>
<td>4</td>
<td>1248</td>
<td>1258</td>
<td>0.041</td>
</tr>
<tr>
<td>5</td>
<td>0721</td>
<td>0731</td>
<td>0.085</td>
</tr>
<tr>
<td>5</td>
<td>0803</td>
<td>0813</td>
<td>0.074</td>
</tr>
<tr>
<td>5</td>
<td>0901</td>
<td>0911</td>
<td>0.103</td>
</tr>
<tr>
<td>5</td>
<td>0945</td>
<td>0954</td>
<td>0.121</td>
</tr>
<tr>
<td>5</td>
<td>1028</td>
<td>1041</td>
<td>0.168</td>
</tr>
<tr>
<td>5</td>
<td>1127</td>
<td>1137</td>
<td>0.172</td>
</tr>
<tr>
<td>5</td>
<td>1212</td>
<td>1223</td>
<td>0.133</td>
</tr>
<tr>
<td>5</td>
<td>1312</td>
<td>1323</td>
<td>0.079</td>
</tr>
<tr>
<td>5</td>
<td>1357</td>
<td>1407</td>
<td>0.068</td>
</tr>
<tr>
<td>5</td>
<td>1454</td>
<td>1500</td>
<td>0.057</td>
</tr>
<tr>
<td>6</td>
<td>0934</td>
<td>0938</td>
<td>0.072</td>
</tr>
<tr>
<td>7</td>
<td>1243</td>
<td>1248</td>
<td>0.064</td>
</tr>
<tr>
<td>7</td>
<td>1326</td>
<td>1334</td>
<td>0.062</td>
</tr>
<tr>
<td>8</td>
<td>0829</td>
<td>0836</td>
<td>0.063</td>
</tr>
<tr>
<td>8</td>
<td>1013</td>
<td>1017</td>
<td>0.065</td>
</tr>
<tr>
<td>8</td>
<td>1308</td>
<td>1318</td>
<td>0.052</td>
</tr>
<tr>
<td>8</td>
<td>1345</td>
<td>1352</td>
<td>0.064</td>
</tr>
<tr>
<td>10</td>
<td>1032</td>
<td>1041</td>
<td>0.062</td>
</tr>
<tr>
<td>10</td>
<td>1114</td>
<td>1124</td>
<td>0.058</td>
</tr>
<tr>
<td>12</td>
<td>1339</td>
<td>1348</td>
<td>0.077</td>
</tr>
<tr>
<td>12</td>
<td>1514</td>
<td>1524</td>
<td>0.080</td>
</tr>
<tr>
<td>12</td>
<td>1602</td>
<td>1616</td>
<td>0.057</td>
</tr>
<tr>
<td>13</td>
<td>1432</td>
<td>1442</td>
<td>0.086</td>
</tr>
<tr>
<td>13</td>
<td>1525</td>
<td>1535</td>
<td>0.573</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>13</td>
<td>1704</td>
<td>1709</td>
<td>0.049</td>
</tr>
<tr>
<td>14</td>
<td>1529</td>
<td>1534</td>
<td>0.102</td>
</tr>
<tr>
<td>14</td>
<td>1652</td>
<td>1706</td>
<td>0.101</td>
</tr>
<tr>
<td>minimum</td>
<td>0.033</td>
<td></td>
<td></td>
</tr>
<tr>
<td>maximum</td>
<td>0.573</td>
<td></td>
<td></td>
</tr>
<tr>
<td>average</td>
<td>0.083</td>
<td></td>
<td></td>
</tr>
<tr>
<td>median</td>
<td>0.064</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 4 – Sensitivity of $\Gamma$ to cloud base temperature and pressure

<table>
<thead>
<tr>
<th></th>
<th>$P_{cb}=900$ hPa</th>
<th>$P_{cb}=950$ hPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_{cb}=5$ °C</td>
<td>$^a\Gamma=1.86$ g m$^{-3}$ km$^{-1}$</td>
<td>$\Gamma=1.92$ g m$^{-3}$ km$^{-1}$</td>
</tr>
<tr>
<td>$T_{cb}=10$ °C</td>
<td>$\Gamma=2.10$ g m$^{-3}$ km$^{-1}$</td>
<td>$\Gamma=2.16$ g m$^{-3}$ km$^{-1}$</td>
</tr>
</tbody>
</table>

$^a$ Evaluated using Eqn. 1, in Albrecht et al. (1990), multiplied by the density of air
Table 5 – Sensitivity of retrieved concentration to $\sigma_g$

<table>
<thead>
<tr>
<th>Geometric Standard Deviation, $\sigma_g$</th>
<th>a, b Relative Concentration Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.05</td>
<td>-0.13</td>
</tr>
<tr>
<td>1.10</td>
<td>-0.11</td>
</tr>
<tr>
<td>1.15</td>
<td>-0.08</td>
</tr>
<tr>
<td>1.20</td>
<td>-0.05</td>
</tr>
<tr>
<td>1.25</td>
<td>-0.03</td>
</tr>
<tr>
<td>1.30</td>
<td>0.00</td>
</tr>
<tr>
<td>1.35</td>
<td>0.03</td>
</tr>
<tr>
<td>1.40</td>
<td>0.05</td>
</tr>
</tbody>
</table>

$^a$ Derived using Eqns. 4a and 4c with the $\Gamma$ derivative is set to zero in Eqn. 4a.

$^b$ The default geometric standard deviation is $\sigma_{g,o} = 1.3$
Table 6 – Spectral dispersions

<table>
<thead>
<tr>
<th>Basis: Observed in-cloud spectra</th>
<th>Basis: Observed in-cloud spectra corrected for instrument broadening</th>
<th>Basis: Parcel model spectra composited into spectra for cloudy mixtures</th>
<th>Basis: Values of $\sigma_{\text{mod}}$ and $&lt;N_{\text{mod}}&gt;$ and Eqn. 7</th>
<th>Basis: Values of $\sigma_{\text{obs}}$ and $&lt;N_{\text{obs}}&gt;$ and Eqn. 7</th>
<th>Basis: Values of $\sigma_{\text{ret}}$ and $&lt;N_{\text{ret}}&gt;$ and Eqn. 7</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.24 ± 0.08 $^b$</td>
<td>0.18 ± 0.07 $^b$</td>
<td>0.14 ± 0.05 $^b$</td>
<td>0.13 ± 0.04 $^b$</td>
<td>0.07 ± 0.02 $^b$</td>
<td>0.24 ± 0.09 $^b$</td>
</tr>
<tr>
<td>0.19 ± 0.06 $^c$</td>
<td>0.16 ± 0.06 $^c$</td>
<td>0.09 ± 0.02 $^c$</td>
<td>0.10 ± 0.02 $^c$</td>
<td>0.07 ± 0.01 $^c$</td>
<td>0.33 ± 0.14 $^c$</td>
</tr>
</tbody>
</table>

$^a$ Instrument broadening was corrected by applying the technique developed by Politovich (1993). A correction factor was derived for each of the 44 analysis intervals. The measured values of $\sigma_D$ were adjusted by subtracting the correction factor in quadrature.

$^b$ Average and standard deviation for all 44 analysis intervals.

$^c$ Average and standard deviation for the 8 analysis intervals with $SS_{\text{eff}} > 0.1 \%$. 


Table A1 – Threshold-diameter tables used for the PCASP and F300

<table>
<thead>
<tr>
<th>Channel</th>
<th>Lower D, ( \mu m )</th>
<th>Upper D, ( \mu m )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.31</td>
<td>0.35</td>
</tr>
<tr>
<td>1</td>
<td>0.35</td>
<td>0.40</td>
</tr>
<tr>
<td>2</td>
<td>0.40</td>
<td>0.46</td>
</tr>
<tr>
<td>3</td>
<td>0.46</td>
<td>0.51</td>
</tr>
<tr>
<td>4</td>
<td>0.51</td>
<td>0.57</td>
</tr>
<tr>
<td>5</td>
<td>0.57</td>
<td>0.64</td>
</tr>
<tr>
<td>6</td>
<td>0.64</td>
<td>0.68</td>
</tr>
<tr>
<td>7</td>
<td>0.68</td>
<td>0.73</td>
</tr>
<tr>
<td>8</td>
<td>0.73</td>
<td>0.80</td>
</tr>
<tr>
<td>9</td>
<td>0.80</td>
<td>1.02</td>
</tr>
<tr>
<td>10</td>
<td>1.02</td>
<td>2.46</td>
</tr>
<tr>
<td>11, 12, 13</td>
<td>2.46</td>
<td>4.69</td>
</tr>
<tr>
<td>14, 15, 16</td>
<td>4.69</td>
<td>6.85</td>
</tr>
<tr>
<td>17, 18, 19</td>
<td>6.85</td>
<td>9.28</td>
</tr>
<tr>
<td>20, 21</td>
<td>9.28</td>
<td>11.3</td>
</tr>
<tr>
<td>22, 23, 24</td>
<td>11.3</td>
<td>12.9</td>
</tr>
<tr>
<td>25, 26, 27, 28</td>
<td>12.9</td>
<td>15.2</td>
</tr>
<tr>
<td>29</td>
<td>15.2</td>
<td>17.7</td>
</tr>
<tr>
<td>30</td>
<td>17.7</td>
<td></td>
</tr>
</tbody>
</table>

Thresholds are an internal electronic representation of the channel boundaries (Cai et al., 2013)
Table B1 – Summary of laboratory-measurements of particle size

<table>
<thead>
<tr>
<th>Test Particle Diameter</th>
<th>Particle Sizing Instrument</th>
<th>a Threshold-diameter Table</th>
<th>b Average Relative Difference</th>
<th>Number of Tests</th>
</tr>
</thead>
<tbody>
<tr>
<td>342, 491 and 707 nm PSL</td>
<td>SMPS</td>
<td>Not Applicable</td>
<td>0.01</td>
<td>59</td>
</tr>
<tr>
<td>342, 491 and 707 nm PSL</td>
<td>F300</td>
<td>Manufacturer</td>
<td>0.09</td>
<td>59</td>
</tr>
<tr>
<td>125, 152, 199, 491 and 707 nm PSL</td>
<td>PCASP</td>
<td>Adjusted</td>
<td>0.03</td>
<td>47</td>
</tr>
</tbody>
</table>

a Thresholds are an internal electronic representation of the channel boundaries (Cai et al., 2013)

b The average relative difference is formulated in terms of the PSL diameter \( D_{i,PSL} \) and the midpoint diameter of the channel with the maximum histogram value. The latter is symbolized \( D_{i,X} \) where “i” is the test number and \( X \) is a place holder for SMPS, F300 or PCASP. In terms of these symbols the average relative difference is \( \langle Y \rangle = n^{-1} \cdot \sum Y_i \) where \( Y_i \) is \( |D_{i,PSL} - D_{i,X}| / D_{i,PSL} \) and the symbol \( “n” \) represents the number of tests.
References -

Albrecht, B., Aerosols, cloud microphysics, and fractional cloudiness, Science, 245, 1227-1230, 1989


Havilcek, L.L., and R.D. Crain, Practical Statistics for the Physical Sciences, American Chemical Society, 1988


Lance, S., Coincidence errors in a cloud droplet probe (CDP) and a cloud and aerosol spectrometer (CAS), and the improved performance of a modified CDP, J. Atmos. Oceanic Technol., 29, 1532-1541, 2012


microphysical and radiative properties of stratocumulus clouds over the southeast Pacific Ocean, Atmos. Chem. Phys., 13, 2541-2562, doi:10.5194/acp-13-2541-2013, 2013

Twomey, S., The nuclei of natural cloud formation part II: The supersaturation in natural clouds and the variation of cloud droplet concentration, Geofisica pura e applicata, 43, 243-249, 1959


81
