

# Comparing parameterized versus measured microphysical properties of tropical convective cloud bases during the ACRIDICON-CHUVA campaign

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## Abstract:

25 The objective of this study is to validate recent parameterizations that were recently developed for satellite retrievals of cloud condensation nuclei supersaturation spectra -  $N_{CCN}(S)$  - at cloud base alongside with more traditional parameterizations connecting  $N_{CCN}(S)$  with cloud base updrafts and drop concentrations. This was based on the HALO aircraft measurements during the ACRIDICON-CHUVA campaign over the Amazon region, which took place in September 2014. The properties of convective clouds were measured with a Cloud Combination Probe (CCP), a  
30 Cloud and Aerosol Spectrometer (CAS-DPOL), and a CCN counter on board the HALO aircraft. An intercomparison of the cloud drop size distributions (DSDs) and the cloud water content (CWC) derived from the different instruments generally shows good agreement within the instrumental uncertainties. To this end, the directly measured

cloud drop concentrations ( $N_d$ ) near cloud base were compared with inferred values based on the measured cloud base updraft velocity ( $W_b$ ) and  $N_{CCN}(S)$  spectra. The measurements of  $N_d$  at cloud base were also compared with drop concentrations ( $N_a$ ) derived on the basis of an adiabatic assumption and obtained from the vertical evolution of cloud drop effective radius ( $r_e$ ) above cloud base. The measurements of  $N_{CCN}(S)$  and  $W_b$  did reproduce the observed  $N_d$  within the measurements uncertainties when using the old (1959) Twomey's parameterization. The agreement between measured and calculated  $N_d$  was only within a factor of 2 when attempting to use cloud base  $S$ , as obtained from the measured  $W_b$ ,  $N_d$  and  $N_{CCN}(S)$ , underscoring the yet unresolved challenge of aircraft measurements of  $S$  in clouds. Importantly, the vertical evolution of  $r_e$  with height reproduced the observation-based nearly adiabatic cloud base drop concentrations,  $N_a$ . The combination of these results provides aircraft observational support for the various components of the satellite retrieved methodology that was recently developed to retrieve  $N_{CCN}(S)$  under the base of convective clouds. This parameterization can now be applied and with the proper qualifications to cloud simulations and satellite retrievals.

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## 1. Introduction

The understanding of cloud formation and its influence on the global hydrological cycle and radiation budget is fundamental for improving weather and climate forecasting models (Ten Hoeve et al., 2011; Jiang and Feingold, 2006; Kohler, 1999; Rosenfeld et al., 2008; Stephens, 1984). The goal of cloud microphysical models is to reproduce atmospheric processes based on physical relationships developed from field experiments and remote sensing observations in different parts of the globe (Silva Dias et al. 2002; Machado et al. 2014; Fan et al. 2014; Rosenfeld et al. 2014b). Data from aircraft probes provide opportunities to validate and improve cloud models and satellite retrievals of cloud microphysical properties.

55 An assessment of the validity of the cloud probe data themselves is essential before the results can be implemented into cloud models. According to previous studies, the number concentration of cloud droplets ( $N_d$ ) expected at cloud base mainly depends on the atmospheric conditions just below cloud base, i.e., updraft wind speed and the supersaturation ( $S$ ) activation spectra of cloud condensation nuclei [ $N_{CCN}(S)$ ] (Pinsky et al., 2012; Reutter et al., 2009; Twomey, 1959). From cloud condensation nuclei counter (CCNC) measurements across a range of supersaturations ( $S$ ), the parameters  $N_0$  and  $k$  are estimated from Twomey's formula (Twomey, 1959):

$$N_{CCN} = N_0 \cdot S^k \quad (1)$$

where  $N_0$  is the cloud condensation nuclei (CCN) concentration at  $S=1\%$  in  $\text{cm}^{-3}$ , and  $k$  is the slope parameter (Twomey, 1959). Equation 1 is an analytical representation of the observational data within the measured range of  $S$ , which in our case represents the observed CCN spectrum from 0.2 to 0.55 %. Note, however, that Eq. 1 does not allow a reliable extrapolation of  $N_{CCN}(S)$  beyond this range (Pöhlker et al., 2016).

65 The parameters  $N_0$  and  $k$  are estimated from data measured below cloud base along with updraft wind speed measurements at cloud base ( $W_b$ ). The values of  $W_b$ ,  $N_0$ , and  $k$  are used for calculating the theoretical cloud droplet concentration from Eq. 2 (Twomey, 1959) below:

$$N_{dT} = 0.88 \cdot N_0^{\frac{2}{k+2}} \cdot (0.07 \cdot W_b^{1.5})^{\frac{k}{k+2}} \quad (2)$$

70 where  $N_{dT}$  are the estimated cloud base drop concentrations in  $\text{cm}^{-3}$ . Here we compare the measured  $N_d$  to  $N_{dT}$  by substituting in Eq. 2 the measured  $N_{CCN}(S)$  in the form of  $N_0$  and  $k$ , along with the measured  $W_b$ .

Equations 1 and 2 are a rather simplistic parameterization. More advanced methods, using the hygroscopicity parameter  $\kappa$  (kappa) are more accurate to represent the CCN spectrum (Pöhlker et al., 2016). However, in this study, using Twomey's parameterization is advantageous, because the CCN measurements were performed within the range of 0.2-0.55 %, where the estimation of the  $N_0$  and  $k$  parameters using Eq. 1 does not incur significant errors in  
75 comparison with more advanced methods (Pöhlker et al., 2016). Furthermore, Twomey's parameterization also allows calculating the effects of updraft wind speed on  $N_{dT}$  as a function of  $N_0$  and  $k$ .

Another approach to estimate the number concentration of CCN that are expected to nucleate as droplets at cloud base is through the use of the  $\kappa$ -Köhler model (Petters and Kreidenweis, 2007). Based on a given dry aerosol particle size distribution (ASD), the  $\kappa$ -Köhler model with prescribed  $W_b$  simulates the expansion and cooling of air as well as  
80 the resulting changes in relative humidity and the related hygroscopic growth of aerosol particles and further condensational growth of cloud droplets. The input to this approach depends strongly on the measured ASD and  $\kappa$  (Reutter et al., 2009).

Measurements of ASD by Passive Cavity Aerosol Spectrometer Probe (PCASP) and Ultra-High-Sensitivity Aerosol Spectrometer (UHSAS) probes were available during the ACRIDICON (Aerosol, Cloud, Precipitation, and  
85 Radiation Interactions and Dynamics of Convective Cloud Systems) - CHUVA (Cloud processes of the main precipitation systems in Brazil: A contribution to cloud resolving modeling and to the GPM [Global Precipitation Measurements]) campaign (Wendisch et al., 2016). However, calculating  $\kappa$  from the combined CCN, PCASP, and UHSAS measurements below cloud resulted in unreasonably low  $\kappa$  values (not shown), which could only be explained by hygroscopic swelling of the aerosols at ambient humidity by a large factor of up to more than two. This  
90 implies that the particles were not completely dried in the intake of the probe, and thus prevents a quantitative assessment of  $\kappa$  based on the PCASP and  $N_{CCN}(S)$ . A possible reason for this behavior in measurements over the Amazon is that the effective hygroscopicity parameters describing water uptake at sub-saturated conditions can be substantially lower than at supersaturated conditions (Mikhailov et al., 2013). The analysis of this effect on the ASD measurements from PCASP and UHSAS below cloud base requires considerable efforts, which are beyond the  
95 scope of this paper. Also, in the case of our flight missions, a major obstacle to the use of the  $\kappa$ -Köhler approach is the fact that measuring the  $N_{CCN}(S)$  spectrum requires a much longer time than the aerosol spectrum with PCASP and/or UHSAS, thus the two measurements are not representing the same aerosol sample. This was evident from the variability of the CCN concentrations measured at fixed  $S$  with one CCNC column, while measuring the  $N_{CCN}(S)$  spectrum with the other column during the flights. The lack of these important analyses prevents the use of  $\kappa$ -Köhler  
100 model estimates for comparison with  $N_d$  measurements from cloud probes in the present study.

An estimation of the cloud base droplet concentrations is also possible via the calculation of the maximum supersaturation ( $S_{max}$ ) at cloud base, relying on the measured  $N_d$  and  $W_b$  according to Eq. 3 (Pinsky et al. 2012) below:

$$S_{max} = C \cdot W_b^{\frac{3}{4}} \cdot N_d^{-\frac{1}{2}} \quad (3)$$

105 where  $C$  is a coefficient that is determined by cloud base temperature and pressure. Since the combination of  $N_{CCN}(S)$  and  $W_b$  determines  $N_d$  and  $S_{max}$ , it is possible to compare the measured and theoretical relationships. Additionally, the estimation of adiabatic cloud droplet concentrations ( $N_d$ ) from measurements of the vertical profile of cloud drop effective radius ( $r_e$ ) is another alternative to evaluate the number of droplets nucleated at cloud base (Freud et al., 2011). The definition of  $r_e$  is:

$$110 \quad r_e = \frac{\int N(r) \cdot r^3 dr}{\int N(r) \cdot r^2 dr} \quad (4)$$

where  $N$  and  $r$  are the droplet concentrations and radii, respectively.

Rosenfeld et al. (2014a) have shown that the effective number concentration of droplets at cloud base ( $N_d^*$ ) can be expressed by a single number, which depends on the effective updraft speed at cloud base ( $W_b^*$ ). To evaluate whether the measured  $N_d^*$  represents the theoretically expected  $N_d^*$  based on the independent measurements of  $N_{CCN}(S)$  and  $W_b$ , it is necessary to find the range of measured  $W_b^*$  and  $N_d^*$  that fulfills best the closure between the measured and indirectly calculated values. Cloud models represent the number of droplets at cloud base by a single number (Pinsky et al., 2012). Therefore, from a set of  $N_d$  measurements at cloud base, an ‘effective’ number of droplets,  $N_d^*$ , can be derived, which represents the measurements for a set of clouds formed in the same thermodynamic condition.

120 The droplet size distribution (DSD) spectrum from clouds, i.e., the DSD variability, depends on the stage of cloud development. After nucleation, the cloud droplets in rising cloud parcels grow with height mainly by condensation. Raindrops start forming when  $r_e$  reaches 13-14  $\mu\text{m}$  and coalescence becomes efficient (Freud and Rosenfeld, 2012; Rosenfeld and Gutman, 1994). Accurate documentation of the vertical evolution of cloud and rain DSDs is essential for analyzing these types of microphysical processes within clouds. Assessing the quality of DSD measurements by the aircraft probes is thus a necessary task. This assessment can be achieved via comparisons between the cloud water content (CWC) calculated from cloud probe DSDs and the direct measurements of CWC with a hot-wire device (CWCh) for cloud penetrations at different heights (Freud et al., 2008; Rosenfeld et al., 2006). This is done in section 3 while accounting for the dependence of the measurement efficiency of the hot-wire on drop size.

130 Three cloud probes measured the DSDs on board the HALO aircraft during the ACRIDICON-CHUVA campaign (Wendisch et al., 2016). In addition, CWC was measured by a King hot-wire probe (King et al. 1978) installed in the Cloud and Aerosol Spectrometer (CAS-DPOL) probe.

Figure 1 illustrates the HALO flight patterns in convective cloud clusters performed in three steps:

- a. Flying below cloud base for measuring  $N_{CCN}(S)$ ;
- b. Flying through cloud base for measuring  $W_b$  and DSD;
- 135 c. Conducting vertical profiles in growing convective towers close to their tops, to avoid precipitation that may fall from above. The cloud penetrations during this phase are made in vertical steps of several hundred meters when possible, from cloud base to the anvils.

The availability of these measurements collected by the same aircraft provides a unique opportunity to compare the data with theoretical predictions and to test the sensitivity of the results to the differences between the measurements by the cloud probes.

This study is novel in several aspects:

- a. It validates the methodology of retrieving the adiabatic cloud drop concentrations  $N_d$  (Freud et al., 2011) from the vertical evolution of  $r_e$  while assuming that  $r_e$  is nearly adiabatic. This is important because it supports the validity of retrieving  $N_d$  from satellite-retrieved vertical profile of  $r_e$  (Rosenfeld et al., 2014a and 2016).
- b. It compares the aircraft-measured  $N_d$  with its parameterization that is based on  $N_{CCN}(S)$  along with the spectrum of updrafts at cloud base weighted by the updraft speed itself,  $W_b^*$ . It is done this way to be compatible with the recently developed methodology of retrieving CCN from satellites by means of retrieving  $N_d$  and  $W_b^*$  (Rosenfeld et al., 2016).
- c. It examines observationally the old Twomey (1959) parameterization of the dependence of  $N_d$  on  $W_b$  (Eq. 2) versus the recent Pinsky et al. (2012) analytical expression for the same relationship (Eq. 3).

These different methodologies are presented in the next sections. Section 2 discusses the instrumentation and database used for this study. Section 3 gives an overview of the cloud probe measurements and discusses consistencies and disagreements between the measurements. Section 4 describes the methodologies applied to compare measurements and model results at cloud base.

## 2. Instrumentation

The HALO flights during the ACRIDICON-CHUVA campaign were performed over the Amazon region, centered on Manaus, during September 2014 under different conditions of aerosol concentration and land cover, as shown in Fig. 2 (from Wendisch et al., 2016). This region was chosen for documenting cloud microstructure and precipitation-forming processes during the dry season with high concentrations of CCN, and to contrast these measurements against cleaner conditions that could be found within flight range, as documented previously (Andreae et al., 2004; Artaxo et al., 2002). Additionally, we made use of the fact that Manaus is located in the central Amazon (3.11 °S; 60.02 °W), and that therefore the aerosol perturbation from the Manaus urban plume may increase CCN concentrations by one to two orders of magnitude above the pristine conditions in the background air (Kuhn et al., 2010). This study is done in collaboration with the Green Ocean Amazon experiment – GoAmazon (Martin et al., 2016), which also addressed the aerosol influences on cloud microphysical properties with special focus on the Manaus urban plume. A comprehensive introduction to airborne instrumentation is given by Wendisch and Brenguier (2013), and in particular of the microphysical instruments involved in this study by Brenguier et al. (2013).

### 2.1 CCN measurements

CCN number concentrations were measured on board HALO during ACRIDICON-CHUVA using a two-column

175 CCNC (CCN-200, Column A and B), a continuous-flow longitudinal-thermal-gradient instrument manufactured by  
Droplet Measurement Technologies (DMT) (Roberts and Nenes, 2005). It measures the CCN number concentration  
as a function of water vapor supersaturation ( $S$ ) at a time resolution of 1 Hz. In the instrument, the sampled aerosol  
particles are exposed to a set supersaturation, and adsorb water depending on their size and chemical composition.  
Those particles that grow to droplets larger than 1  $\mu\text{m}$  in diameter are counted as CCN at that  $S$ . The instrument was  
calibrated between flights following Rose et al. (2008). The estimated uncertainties for CCN number concentration  
180 is about 20 % (10 %) on average for large (small) concentrations. In addition, the uncertainty on supersaturation  
values is 10 % on average.

Sample air for the aerosol measurements was obtained from two different inlets: (i) the HALO aerosol submicron  
inlet (HASI), and (ii) the HALO counterflow virtual impactor (HALO-CVI) (Wendisch et al., 2016). The CCN-200  
provides the possibility to measure in parallel from both inlets or at two different values of  $S$ . In this study, only the  
185 aerosol measurements from the HASI inlet have been used. The measurements were done with one column at a  
constant  $S=0.55\%$ , while the other was cycling  $S$  between 0.2 and 0.55 % with steps every 100 seconds.

## 2.2. Cloud probe measurements

Three cloud probes were operated on board HALO during the measurements in the ACRIDICON-CHUVA  
190 campaign. This study focuses on the CAS-DPOL and CCP-CDP probes. The third probe, NIXE-CAS-DPOL was of  
identical type as CAS-DPOL and is thus not used in this study. The probes' range of measurements is shown in  
Table 1. In this study, cloud particle concentrations are counted at diameters larger than 3  $\mu\text{m}$  to avoid  
measurements of haze droplets. This is also in accordance with the similar lower limits of the bins sizes of the CCP-  
CDP. Details of the cloud probe measurement characteristics are described in the following sections (see also  
195 Brenguier et al., 2013).

### 2.2.1 CCP-CDP and CCP-CIP measurements

The Cloud Combination Probe (CCP) combines two detectors, the Cloud Droplet Probe (CDP) and the greyscale  
Cloud Imaging Probe (CIPgs). The CDP detects forward scattered laser light from cloud particles as they pass  
200 through the CDP detection area (Lance et al., 2010) and represents an advanced version of the Forward Scattering  
Spectrometer Probe (FSSP) (Baumgardner et al., 1985; Dye and Baumgardner, 1984; Korolev et al., 1985; Wendisch  
et al., 1996). The CIPgs records 2-D shadow-cast images of cloud elements that cross the CIPgs detection region.  
The overall particle detection size range is 2 to 960  $\mu\text{m}$  when measuring with the CCP. The highest temporal  
resolution of the CCP measurements is limited to 1Hz. Recent findings concerning the measurement uncertainties of  
205 the underwing cloud probes at the comparatively high HALO flight velocities (well above 170  $\text{m s}^{-1}$ ) provide  
correction procedures to be applied to the measured raw data to further improve the data quality of the ambient  
cloud particle number concentrations (Weigel et al., 2016). The robust performance of the specific CCP instrument  
used in this study, even under extreme conditions, was demonstrated by earlier investigations in tropical convective  
outflow (Frey et al., 2011), Polar Stratospheric Clouds (PSC) (Molleker et al., 2014), and low-level mixed-phase  
210 clouds in the Arctic (Klingebiel et al., 2015).

For the CDP sample area of 0.27 mm<sup>2</sup>, an uncertainty of about 10% was considered (Molleker et al., 2014). The uncertainty  $\pm 0.03$  mm<sup>2</sup> results from repeated measurements. Unless there is no massive manipulation/disarrangement of the CDP's optics or a detectable aging of the laser diode, the sample area remains stable even if the instrument experiences regular handling during, e.g., field campaign operations. Given the uncertainty of the sample area, the Probe Air Speed (PAS), particle losses, deviations and maybe coincidence (not negligible, but likely not a significant issue), the uncertainty in cloud droplet concentration ranges below 20% and likely approaches or exceeds 20% only in cases of tight curve maneuvers as this might be the most prominent case when the "collecting angle" comes into play. For the flight pattern adopted during vertical profiling of clouds (where cloud penetrations were performed in straight and level flight) the uncertainty of the number concentration for CCP-CDP is 10%.

### 2.2.2 CAS-DPOL measurements

The CAS-DPOL measures particle size distributions between 0.5 and 50  $\mu\text{m}$  at 1-Hz time resolution (Baumgardner et al., 2001). Its measurement principle is developed based on the FSSP-300 (Baumgardner et al., 1985, Korolev et al., 1985), which has been used previously to study the particle size range in ice clouds (Voigt et al., 2010, 2011; Schumann et al., 2011; Jeßberger et al., 2013). The intensity of forward scattered light in the angular range of 4 – 12° is detected and sorted into 30 size bins. Assuming Mie scattering theory, additional binning into 15 size bins is employed to rule out ambiguities. Polarized backward scattered light is detected to investigate the sphericity and phase of the particles (Baumgardner et al., 2005; Gayet et al., 2012; Järvinen et al., 2016). Number concentrations are derived using the probe air speed measured by the probe. The distribution of time intervals between single particles, recorded for the first 290 particles in each second, did not provide indications of droplet coincidence up to a time resolution of 0.8  $\mu\text{s}$  or a number concentration of 2200 cm<sup>-3</sup>. After the campaign, the sampling area (SA) which is used to derive the number concentration of particles was characterized by a high-resolution scan with a droplet generator. For this, 250 water droplets of a known, quasi-constant size of about 40  $\mu\text{m}$  were dropped at and around the sensitive region perpendicular to the laser beam. The resolution of the droplet generator scan was 25  $\mu\text{m}$  perpendicular to the laser beam and 50  $\mu\text{m}$  along the laser beam. According to the scan, the area of the measured SA for particle diameters above 3  $\mu\text{m}$  was 0.27 mm<sup>2</sup>, which is 8% higher than the initially reported SA by the manufacturer. The fringe of the area, a region where particles are counted but with low efficiency was about 0.032 mm<sup>2</sup> which represents an uncertainty of 15% of the total SA. Additionally, we estimate an uncertainty of the particle velocity in the CAS sampling tube of 15%, taking into account that particle velocities in the sampling tube may be slowed down or accelerated compared to open path instruments or the Pitot tube velocities at the CAS. This results in a combined uncertainty of the number concentration of 21%.

Calibrations with glass beads of four different sizes (2, 5, 20 and 42  $\mu\text{m}$ ) were performed between the flights to monitor the stability of the size bin classification. Differences in the refractive index can be accounted for using the method of Rosenberg et al. (2012). The size calibration was stable over the whole campaign. For the purpose of this study, mainly the effective diameter range between 10 and 26  $\mu\text{m}$  was evaluated, which employed mainly the lowest amplifier gain stage. For particles up to 20  $\mu\text{m}$  in size, the calibration did not show any size deviations from the

250 expected values. Larger particles with diameters  $> 40 \mu\text{m}$  were shifted towards lower sizes by about  $5 \mu\text{m}$ . We therefore estimate an uncertainty in particle size for particle diameters above  $40 \mu\text{m}$  on the order of 13 to 15%, and less for smaller particles. The instrument had been installed previously on HALO and the DLR Falcon aircraft during the ML-CIRRUS (The Midlatitude Cirrus)[Voigt et al., 2016], ACCESS-II (Alternative Fuel Effects on Contrails and Cruise Emissions) [Moore et al., 2017], and DACCIWA (Dynamics-Aerosol-Chemistry-Cloud Interactions in West Africa) [Knippertz et al., 2015] campaigns.

### 255 **2.3 Hot-wire CWC measurements**

The hot-wire instrument is a King Probe type device that measures the bulk liquid water content (LWC) from  $0.01$  to  $3 \text{ g m}^{-3}$  in the droplet diameter range of  $5$  to  $50 \mu\text{m}$  by detecting the power (current) required to maintain a heated wire at a constant temperature of  $125^\circ\text{C}$ . The sensitivity of the instrument is reduced for droplets below  $10 \mu\text{m}$ , since smaller particles follow more closely the streamlines around the hot-wire. The instrument was mounted on the CAS-DPOL probe. The accuracy of the King Probe LWC measurement is estimated to be  $5 \%$  at  $1 \text{ g m}^{-3}$  and decreases down to  $16 \%$  at  $0.2 \text{ g m}^{-3}$ , with a sensitivity of  $0.02 \text{ g m}^{-3}$  (King et al., 1978). For this study, mainly CWC values in the range up to  $1 \text{ g m}^{-3}$  were used.

### 265 **2.4 Vertical wind speed measurements**

The HALO aircraft was equipped with a new meteorological sensor system (Basic HALO Measurement And Sensor System - BAHAMAS) located at the nose of the aircraft (Wendisch et al., 2016). Measurements of updraft speeds during cloud base penetrations during the ACRIDICON-CHUVA campaign showed maximum vertical wind speeds in the range of  $5 \text{ m s}^{-1}$ . In these conditions, the uncertainties of  $W$  measurements are  $0.3 \text{ m s}^{-1}$  (Mallaun et al., 2015).

## 270 **3. Cloud probe intercomparison**

### **3.1 Method**

275 The validation of convective cloud parameterizations requires reliable cloud probe measurements. In this section, we discuss quantitatively the differences in estimated and directly measured CWC and DSDs of the two cloud probes CAS-DPOL and CCP-CDP as well as the hot-wire instrument.

For comparisons between the CWC estimated from the cloud probe DSDs and hot-wire measurements ( $CWCh$ ), we distinguish between spectra that are dominated by condensational growth, and spectra where coalescence becomes important as well. These spectra are separated by the threshold of  $r_e$  for significant coalescence, which varies as a function of the drizzle water content (DWC) for 1 second cloud passes (Freud and Rosenfeld, 2012). In addition, droplets with diameters  $< 10 \mu\text{m}$  are captured less efficiently by the hot-wire probe, resulting in an underestimation of  $CWCh$ . The hot-wire device was installed on the CAS-DPOL probe; therefore a better statistical agreement is expected for this probe in comparison with the CCP-CDP. The CCP-CDP was mounted on the other wing, about  $15 \text{ m}$  away from the hot-wire device (Voigt et al., 2016; Wendisch et al., 2016). Only cloud passes at temperatures greater than  $0^\circ\text{C}$  are considered in this intercomparison, to avoid uncertainties of the measurement due to freezing of



droplets.

### 3.2 CWC comparison between cloud probe and hot wire measurements

290 Comparison of different techniques of cloud water content measurements are challenging because of the individual instrumental differences, like time resolution, dependence of sensitivity on size, and due to the characteristics of their target of interest, i.e., inhomogeneous, turbulent convective cloud.

For this study we use the hot-wire instrument as a reference to the scattering spectrometer probes, since its total water content is derived from a smaller set of physical parameters with an overall maximum uncertainty of 16% as compared to ~ 30% uncertainty when derived from DSDs.

295 The calculation of CWC is performed separately from CAS-DPOL and CCP-CDP probe droplet concentrations as follows:

$$CWC = \frac{4\pi}{3} \rho \int N_d(r) r^3 dr \quad (5)$$

where  $N_d$  is the droplet concentration in  $\text{m}^{-3}$ ,  $r$  the droplet radius in m and  $\rho$  is the water density ( $1 \text{ g cm}^{-3}$ ). The calculation of DWC is done similar to CWC but with different particle size ranges from CCP-CIP measurements.

300 The DSDs from CCP-CDP and CAS-DPOL are used to calculate the CWC, defined here as the mass of the drops integrated over the diameter range of 3–50  $\mu\text{m}$ . Similarly, DSDs from CCP-CIP are used to calculate the DWC, defined here as the mass of the drops integrated over the diameter range of 75–250  $\mu\text{m}$  (Freud and Rosenfeld, 2012).

Figure 3 shows the dependency of calculated  $r_e$  as a function of altitude for cloud passes during flights over different conditions of aerosol concentrations (AC13 - very polluted, AC18- polluted, and AC19 – clean). The probability of

305 rain due to collision and coalescence processes are indicated with dashed lines. It is assumed that rain formation starts when calculated DWC exceeds  $0.01 \text{ g m}^{-3}$  (Freud and Rosenfeld, 2012). Overall, the figure shows that  $r_e$

values increase with altitude. In addition, it shows the effects of aerosol loading, which in higher concentration nucleate a larger number of droplets at cloud base, which grow slower as a function of height via condensation.

310 Also, for  $r_e$  values  $< 9 \mu\text{m}$  the probability of coalescence of droplets is very small and it starts to be significant only for  $r_e > 11 \mu\text{m}$ . There is little concern that raindrops precipitate from above when flying near the tops of growing convective clouds (as illustrated at Fig. 1).

The comparison of CWC estimated from the cloud probe data and  $CWCh$  measured with the hot-wire was performed as a function of  $r_e$ , because the measurement efficiency of the hot-wire probe depends on drop size. This

315 type of analysis also provides information about the differences between the two cloud probes regarding the estimated CWCs. Strapp et al. (2003) show that large differences between actual CWC and hot-wire measurements occur when larger drops ( $\sim r > 20 \mu\text{m}$ ) contribute to the cloud water content above  $1 \text{ g m}^{-3}$ . We therefore limit our

analysis to the effective diameter range of  $5 \mu\text{m} < r_e < 13 \mu\text{m}$  and compare  $CWCh$  with CWC estimated from the cloud probe DSD only for CWC up to  $1 \text{ g m}^{-3}$ . The comparison between the mean CWC estimated from the cloud

probe DSDs and mean  $CWCh$  is shown as a function of  $r_e$  in Fig. 4. The ratio between the  $CWCh$  from the hot-wire

320 measurements and the probe estimates ( $CWC_r$ ) is also shown (in red color).

The mean values of CWC estimated from the probes from flights AC08 to AC20 (AC07 had no hot-wire CWC data) and altitudes between 600 m and 5,000 m generally show an increase with increasing  $r_e$ . The CWC uncertainty

325 calculated with CAS-DPOL (CCP-CDP) DSDs is about 22% (10 %) for all measurements. In addition, the uncertainty associated with  $r_e$  calculations with CAS-DPOL (CCP-CDP) DSDs is about 14 % (9 %). Within their statistical variability, the CAS-DPOL CWC agrees well with the hot-wire CWCh over the whole effective radius range (upper panel). The CWC<sub>r</sub> for CAS-DPOL (CCP-CDP) is around  $1 \pm 0.1$  ( $0.8 \pm 0.05$ ) for almost all  $r_e$  sizes. The comparisons of the CWCh with the CWC estimated from the CCP-CDP probe (lower panel) shows that the CCP-CDP is systematically higher by about 21%. The difference is larger than the standard deviation of the individual measurements. The overall systematic differences (mean of the ratio) in the cloud probe CWC in comparison to CWCh are  $0.04 \text{ g m}^{-3}$  (6% in percentage) for CAS-DPOL and  $0.11 \text{ g m}^{-3}$  (21% in percentage) for CCP-CDP higher than the hot-wire measurements. However, considering the uncertainty of the measurements, all three CWC measurements agree within the uncertainty range (16% and 30%).

330 In summary, the CWCh from the hot-wire agrees better with the CWC derived from CAS-DPOL DSDs. The fact that the CCP-CDP was mounted on the opposite wing while the measurements were performed in very inhomogeneous conditions may account for some of the larger spread between CCP-CDP and hot-wire than between CAS-DPOL and hot-wire (e.g., in  $r_e$ ), but it cannot explain the systematic offset of the CCP-CDP. In the next subsection we discuss input parameters for the CWC estimated from the cloud probes, like number concentration and size to find an explanation for the observed differences.

### 340 3.3 Comparing cloud probe $N_d$ and DSDs

Figure 5 shows the mean  $N_d$  values measured by CAS-DPOL and CCP-CDP (solid line) and the systematic uncertainties of the measurements (dashed lines) as a function of  $r_e$  for values greater than  $5 \mu\text{m}$  (left panel) and the standard deviation of the two cloud probe  $N_d$  measurements (right panel). The data are the same as used for the hot-wire intercomparison. Both probes measure a decreasing number concentration with increasing effective radius and CWC at greater heights above cloud base. This is related to the increasing extent of mixing and coalescence processes with height in the cloud. Therefore, a reduced number of larger droplets contribute to the enhanced CWC at larger  $r_e$ . In general, the CAS-DPOL mean  $N_d$  agrees well (difference lower than 1 %) with the mean  $N_d$  of CCP-CDP for effective radii between  $7$  and  $11 \mu\text{m}$ . Statistically significant differences are observed for  $r_e$  smaller than  $7 \mu\text{m}$  and above  $11 \mu\text{m}$ . Both probes have similar standard deviation (STDE) for different  $r_e$  sizes. The STDE decreases with increasing  $r_e$ , varying from  $\sim 20 \text{ cm}^{-3}$  to  $\sim 10 \text{ cm}^{-3}$ .

350 The two  $N_d$  measurements agree within the combined statistical variability and the systematic uncertainties of the two probe measurements (21% for CAS-DPOL and 10% for CCP-CDP). However, in order to explain the difference in CWC, we point towards the difference in the mean droplet number at  $r_e > 11 \mu\text{m}$ . Lower number concentrations of the CAS-DPOL at larger  $r_e$  may be related to the shift in droplet radii for particles above  $40 \mu\text{m}$  to smaller sizes, which shifts the effective radius and the CWC to smaller  $r_e$  and smaller CWC. On the other hand, the difference in the size binning of the two probes may artificially shift particles from higher sizes to lower sizes just by the choice of the bin boundaries. For the CAS-DPOL, larger bin sizes were chosen in order to avoid ambiguities based on Mie-Lorenz theory.

The differences in  $N_d$  at larger  $r_e$  correspond to the enhanced CWC in Fig. 4 and may explain most of the differences

360 in CWC between the probes. The higher number concentration at  $r_e < 7 \mu\text{m}$  may be explained by the higher sensitivity and lower cutoff of the CAS-DPOL at smaller sizes. The instrument was built to particularly measure the full spectrum of aerosol and cloud particles in the size range where aerosols are activated into cloud droplets.

Figure 6a-d shows the mean droplet concentration and CWC as a function of droplet diameter from the cloud probes. The distributions are shown for four different effective radii to give an impression of the evolution of particle size and CWC with altitude for the two cloud probes. For  $r_e$  between 5 and 6  $\mu\text{m}$  and 8 and 9  $\mu\text{m}$  (Figures 6a-b), where collision and coalescence processes are negligible (see Fig. 3), the CCP-CDP DSDs are somewhat below the CAS-DPOL DSDs, revealing an enhanced sensitivity of the CAS-DPOL for smaller particles. For larger  $r_e$  (Figures 6c-d), where coalescence starts and raindrops may be present, the CCP-CDP shows slightly larger droplet concentrations and CWC for diameters  $> 15 \mu\text{m}$  in comparison to CAS-DPOL. This may be related to larger droplets that enter the open path instrument sampling area of the CCP-CDP more easily than the closed path sampling area of the CAS-DPOL by falling vertically into the measurement area.

370 These results suggest that CAS-DPOL and CCP-CDP generally measure similar droplet concentrations in the size range between 3-50  $\mu\text{m}$ . The observed deviations between the probes could be caused by different inlet configurations or measurement principles of the two probes, each with individual advantages depending on the measurement target and related size range. However, the differences in DSDs are within the uncertainties of the measurement and show a much better agreement than earlier measurements under similar conditions (Lance et al., 2012; Rosenberg et al., 2012).

#### 4. Methodology

380 The reliability of the cloud probe measurements shown in the previous section provides the capability to perform the validation of convective cloud parameterizations for the Amazon region, and these analyses are performed through the following four steps. Section 4.1 presents the analyses of CCN measurements below cloud base. Assuming the relation between  $N_{\text{CCN}}$  and  $S$  is given by Eq. 1, the parameters  $N_0$  and the slope  $k$  are calculated from the measurements below cloud base. Section 4.2 describes the estimation of maximum  $S$  at cloud base ( $S_{\text{max}}$ ) based on the measured  $N_d$  and  $W_b$  there. The co-variability of  $N_d$  and  $W_b$  is used to estimate the CCN concentration ( $N_{d\text{CCN}}$ ) by calculating  $S_{\text{max}}$  according to Eq. 1. This is repeated for the two  $N_d$  spectra that were obtained from the two cloud droplet probes. In addition,  $N_d$  is estimated by application of the measured  $W_b$  spectrum to Eq. 2 and comparing against the directly measured  $N_d$  from the two cloud probes. Section 4.3 outlines the methodology of calculating the effective number of droplets at cloud base from cloud probe measurements ( $N_d^*$ ). This is done using theoretical considerations based on the estimated values of  $N_{dT}$  and  $N_{d\text{CCN}}$  at cloud base ( $N_{dT}^*$  and  $N_{d\text{CCN}}^*$ , respectively). The exact definitions of all parameters are provided in Section 4.2. Section 4.4 explains the calculation of the estimated adiabatic cloud droplet concentration ( $N_a$ ), as obtained from the measured vertical profile of cloud drop size distributions.

##### 395 4.1 CCN measurements below cloud base as a function of $S$

The measurements of  $N_{\text{CCN}}$  and  $S$  can be parameterized by Eq. 1 and provide  $N_0$  and  $k$  (Pruppacher et al., 1998). The

typical values of  $N_0$  are about  $100 \text{ cm}^{-3}$  for pristine conditions, and range from  $500 \text{ cm}^{-3}$  to several thousand  $\text{cm}^{-3}$  for polluted continental regions at different levels of aerosol loading. The values of the slope parameter  $k$  vary from about 0.3 to 1 in clean and polluted air, respectively (Andreae, 2009).

400 Two types of CCN measurements were performed: (i) measuring CCN concentration at fixed  $S$  ( $\sim 0.55\%$ ) [hereafter referred to as  $S_1$  with the corresponding CCN concentration referred as  $CCN_1$ ] and (ii) measuring CCN concentration at variable  $S$  (ranging from 0.2 % to 0.55 %) [hereafter referred to  $S_2$  with the corresponding CCN concentration referred as  $CCN_2$ ]. Since the  $CCN_2$  measurements were performed at varying  $S_2$  (generally modified every 100 seconds during the flights; hereafter referred as time step), the mean values of these measurements for  
405 each time step are used to calculate the  $N_0$  and  $k$  parameters in Eq. 1. The flight period of measurements below cloud base in a specific region consisted of several CCN time steps and covered at least one full  $N_{CCN}(S)$  spectrum, and is defined as a group of measurements (hereafter referred as a group).

To achieve accurate measurements of  $CCN_2$  as a function of  $S_2$ , a weighting factor calculated from the  $CCN_1$  measurements is applied, as specified in the steps below. Because  $CCN_1$  measures at a fixed supersaturation ( $S_1$ ), its  
410 variability is caused only by changes of total CCN concentration (from aerosol loading) along the flight track (assuming constant size distribution and composition during the measurement group). This is used to correct the  $N_{CCN}(S)$  as measured by  $CCN_2$  for these changes of total concentration. The procedure for this analysis is:

1. The mean values of  $S_1$ ,  $S_2$ ,  $CCN_1$  and  $CCN_2$  measurements ( $mS_1$ ,  $mS_2$ ,  $mCCN_1$  and  $mCCN_2$ , respectively) are calculated for each time step below cloud base;
- 415 2. A factor of aerosol loading ( $FA$ ) for measurements during a full cycle of  $S$  is calculated as follows:

$$FA = \frac{mCCN_1}{TmCCN_1}$$

where  $TmCCN_1$  is the mean of all  $CCN_1$  measurements for the group of  $S$  cycling.  $FA$  provides the deviation of aerosol concentration from the mean for a specific time step in the group;

3. The  $mCCN_2$  values for each group are weighted by  $FA$  generating normalized  $mCCN_2$  values ( $NCCN_2 = mCCN_2 / FA$ ). Then, the  $NCCN_2$  are used in combination with  $mS_2$  to fit a power-law-function equation for  
420 each group of measurements. From this fit, the values of the parameters  $N_0$  and  $k$  in the Twomey equation ( $N_{CCN} = N_0 \cdot S^k$ ) are obtained.

#### 4.2. Estimating $S_{max}$ , $N_{dCCN}$ , and $N_{dT}$

The number of CCN that nucleate into cloud droplets ( $N_d$ ) reaches its maximum value near the  $S_{max}$  height in the cloud (Pinsky et al., 2012). This level is observed between cloud base and a height up to a few tens of meters above  
425 it. The value of  $S_{max}$  can be estimated from Eq. 3 based on the vertical velocity at cloud base and on  $N_d$  as measured with the cloud probes CCP-CDP and CAS-DPOL ( $N_{cdp}$  and  $N_{cas}$ , respectively). Therefore, the estimated  $S_{max}$  near cloud base can be used in Eq. 1, producing the  $N_{dCCN}$  estimates to hopefully achieve a closure for  $N_d$  measurements at cloud base.

430 The  $N_0$  and  $k$  values that were calculated from measurements below cloud base (as described in Section 4.1) are substituted in Eq. 1 and Eq. 2 for calculating  $N_{dCCN}$  and  $N_{dT}$ , respectively. The comparisons between  $N_{dCCN}$ ,  $N_{dT}$  and

435  $N_d$  from the cloud probes are discussed in Section 5.2. Measurements of  $N_d$  for each probe are considered only for concentrations  $\geq 20$  droplets per cubic centimeter, to focus on the convective elements and avoid highly mixed and dissipating portions of the clouds. The time and distance differences that were allowed between the measurements below cloud base and at cloud base have maximum values of 1 hour and 30 km, respectively. With this consideration, we assume that the  $N_d$  measurements at cloud base pertain to the same region as the CCN measurements below cloud base.

440 According to Twomey (1959), the  $N_d$  that should be observed at cloud base increases with  $W_b$  (assuming a constant CCN concentration; see Eq. 2). However, at cloud base the variability of  $W_b$  and  $N_d$  measurements is high due to air turbulence. Since a cloud parcel moves as an eddy with a local  $W_b$  that produces a given  $N_d$  at cloud base, its continued movement as a turbulent eddy within the cloud adds a large random component to the individual realizations of  $W_b$  for a given  $N_d$ . These turbulent characteristics greatly reduce the confidence that a given measured  $W_b$  within cloud has produced the corresponding measured  $N_d$ , and therefore, these measurements are often not well correlated. A suitable method to analyze the relationship between  $W_b$  and  $N_d$  measurements is the ‘probability matching method’ (PMM) (Haddad and Rosenfeld, 1997), which requires that the two related variables will be increasing monotonically with each other. For a set of measurements of  $W_b$  and  $N_d$  at cloud base, it is expected that larger  $W_b$  would produce larger  $N_d$  for a given  $N_{CCN}(S)$ . Therefore, it is assumed also that  $N_d$  is produced uniquely by  $W_b$  for a given  $N_{CCN}(S)$  spectrum as calculated from the measurements below cloud base. It is further assumed that entrainment does not change systematically with  $W_b$  in a way that would reverse the monotonic increase of  $W_b$  with  $N_{CCN}(S)$ . In a PMM analysis, the same percentiles of updrafts are matched to the same percentiles of  $N_d$  (or  $N_{dCCN}$  and  $N_{dT}$ ). As  $N_d$  must be produced by positive updrafts (Eq. 2), negative (positive) values of  $W_b$  are associated with lower (higher)  $N_d$ . This procedure allows identifying the role of  $W_b$  (positive) in producing  $N_d$  in a set of cloud base measurements. The results of PMM analysis from cloud probes  $N_d$  versus  $W_b$ , and for estimated  $N_{dCCN}$  with  $N_{dT}$  are discussed in Section 5.2.1.

455

### 4.3. Estimating $W_b^*$ , $N_d^*$ , $N_{dT}^*$ and $N_{dCCN}^*$

The formulation of an effective updraft speed at cloud base ( $W_b^*$ ) is a useful approximation of the updraft spectrum (Rosenfeld et al., 2014a; Zheng et al., 2015).  $W_b^*$  and  $N_d^*$  are given in Eqs. (6) and (7):

$$W_b^* = \frac{\int W_b^2}{\int W_b} \quad ; \text{where } W_b > 0 \quad (6)$$

$$N_d^* = N_d[\text{percentile}(W_b^*)] \quad (7)$$

460

where  $N_d^*$  represents the spectrum of  $N_d$  at cloud base that matches the same percentile of  $W_b^*$ . Figure 7 shows an illustration and example of the estimated value of  $W_b^*$  and  $N_d^*$  from the CCP-CDP probe for flight AC17. In this case the calculated  $W_b^*$  has a value of  $1.83 \text{ m s}^{-1}$ , which represents the 86<sup>th</sup> percentile of total measurements at cloud base when sorted by  $W_b$  measurements, including negative values. The corresponding percentile of  $N_d^*$  (when sorted by  $N_d$ ) in this case is  $1207 \text{ cm}^{-3}$ . Another approach for  $N_d$  retrieval is the calculation of  $N_{dT}^*$  considering  $W_b^*$  as the

465

updraft wind speed in Eq. 2. In addition,  $S_{max}$  can be estimated by applying the calculated values of  $W_b^*$  and  $N_d^*$  to Eq. 3. Then, applying the obtained  $S_{max}$  to Eq. 1 yields  $N_{dCCN^*}$ . The values of the calculated  $N_{dT^*}$  and  $N_{dCCN^*}$  in this case are  $1,175 \text{ cm}^{-3}$  and  $915 \text{ cm}^{-3}$ , respectively.

#### 470 **4.4. Estimating $N_a$**

Another approach for estimating  $N_d$  is through the calculation of the adiabatic cloud droplet number concentration,  $N_a$  (Freud et al., 2011). The  $N_a$  is calculated from CWC and the mean volume droplet mass ( $M_v$ ) calculations from the cloud probe DSDs obtained during the cloud profiling measurements. This behavior is the outcome of the almost completely inhomogeneous mixing behavior of the clouds with the ambient air (Burnet and Brenguier, 2007; Freud et al., 2011). Recently, Beals et al. (2015) wrote that their "*measurements reveal that turbulent clouds are inhomogeneous, with sharp transitions between cloud and clear air properties persisting to dissipative scales (<1 centimeter). The local droplet size distribution fluctuates strongly in number density but with a nearly unchanging mean droplet diameter*". The dominance of inhomogeneous mixing diminishes when the drops become very large ( $r_e > 15 \text{ }\mu\text{m}$ ) and their evaporation rate becomes more comparable to the mixing rate. This is most evident in those cloud passes where CWC is greater than 25 % of the adiabatic CWC (Freud et al., 2011). The measurements during cloud profiling flights were aimed at penetrating the tops of growing convective towers (as shown at Fig. 1). This was done successfully in the data selected for analysis, as verified by examination of videos recorded by the cockpit camera of HALO. The cloud penetrations occurred mainly near the tops of growing convective cumulus, where mixing is expected to be rather inhomogeneous and little precipitation can fall from above. The validity of this expectation will affect the agreement between  $N_d$  and  $N_a$ . The  $N_a$  is calculated from the slope of CWC and  $M_v$  measurements and provides an estimate of  $N_d^*$  near cloud base. However, this methodology does not account for cloud mixing losses from droplet evaporation and the  $N_a$  estimates commonly overestimate the expected  $N_d$  by 30 % (Freud et al., 2011). Therefore, in calculating  $N_a$  we applied this 30 % correction.

### 490 **5. Results**

#### **5.1 CCN measurements below cloud base**

The estimation of the  $N_0$  and  $k$  parameters in Eq. 1 is made from CCN and  $S$  measurements below cloud base. Figure 8 illustrates CCN and  $S$  measurements below cloud base for flight AC17 over a deforested region in the central Amazon. The cloud base was located at a height of about 2,300 m. The values of  $S_1$  were constant at  $\sim 0.55 \%$  and the values of  $S_2$  ranged from 0.2 % to 0.55 %. During these measurements,  $CCN_1$  showed higher values than  $CCN_2$ , which is in agreement with its larger  $S$ , and the difference between  $CCN_1$  and  $CCN_2$  increased with decreasing  $S_2$  (e.g., at time  $\sim 19:45$  UTC, where  $CCN_2$  values are around  $300 \text{ cm}^{-3}$  and  $CCN_1$  values are around  $700 \text{ cm}^{-3}$ ). The  $mCCN_1$ ,  $mCCN_2$ , and  $NCCN_2$  for this group of measurements are shown in Fig. 9. The average measurements of  $CCN_1$  ( $TmCCN_1$ ) assumed to correct the  $CCN_2$  for aerosol load ( $FA$ ) presented a standard deviation of 14 %, indicating a small impact on the parameterization proposed to fit the Twomey equation (Eq. 1). The power fit equation from  $NCCN_2$  and  $mS_2$  measurements is shown and the values of  $N_0$  and  $k$  are  $1015 \text{ cm}^{-3}$  and 0.54, respectively.

This procedure was applied to all cloud profiling flights with measurements of  $N_{CCN}(S)$  with variable  $S$  below cloud base. The  $N_0$  and  $k$  slope parameters for all groups of measurements during the campaign are shown in Fig. 10. The measurements show that for the less polluted conditions, the values of  $N_0$  ( $k$  slope) are near 1000 (0.5), while for more polluted conditions, values of  $N_0$  ( $k$  slope) greater than 2000 (0.9) are observed. Additionally, the correlation coefficient values for almost all power fit equations are around 0.9. The calculated  $N_{CCN}(S)$  errors for these flight segments are a function of the measured particle number, such that the error is 10% of  $N_{CCN}(S)$  for large concentrations and the mean of the error is around 20% of  $N_{CCN}(S)$ . The estimated standard error (STDE) for the  $N_0$  and  $k$  parameters and CCN estimates were calculated (as described in Appendix B) for each flight segment and are shown in Table 2. The table shows that the STDE associated with the Twomey equation fit is about 5% for the  $N_0$  and  $k$  parameters. The changes in the air mass assumed to correct the  $CCN_2$  for  $FA$  during the flight segments were up to 24 % for all flights. When the cloud segment compared with this data are not at exactly the same location as where the measurements were performed, the mean (i.e.  $TmCCN_1$ ) is a good measure for this comparison. The standard error was used for the error propagation calculations and the resulting error in  $N_{CCN}(S)$  is 15 % of the  $N_{CCN}(S)$  estimates on average. The resulting error of  $N_0$  ( $k$  slope) was also calculated and is 23 % (20 %) of the  $N_0$  ( $k$ ) values on average, associated to the Twomey equation fit and the  $N_{CCN}(S)$  error.

For some flights, the values estimated for the  $N_0$  and  $k$  parameters of Eq. 1 are similar to what was found by Pöhlker et al. (2016) for ground measurements at the Amazon Tall Tower Observatory (ATTO) site ( $N_0 = 1469 \pm 78$  and  $k = 0.36 \pm 0.06$ ) during the dry season in the Amazon. However, in the majority of the cases  $N_0$  and  $k$  are twice or three times greater than the values from Pöhlker et al. (2016). These differences are probably related to flying selectively to areas that had high aerosol concentrations to contrast the cloud behavior with the flights with low aerosol concentrations, as shown in Fig. 2. The high CCN measured in this study are more similar to previous aircraft measurements in smoky conditions over the Amazon (Andreae et al., 2004; Freud et al., 2008) than to the values observed at ATTO.

## 5.2 Comparing estimated with measured $N_d$ near cloud base

Cloud base drop concentrations obtained in several different ways were compared. Appendix C summarizes the measurements and theoretical calculations at cloud base. Agreement between these different estimates constitutes a closure. Section 5.2.1 discusses comparisons between individual cloud probe  $N_d$  measurements with the corresponding theoretical estimations of  $N_{dT}$  and  $N_{dCCN}$ . Section 5.2.2 describes the comparisons between estimated  $N_d^*$ ,  $N_{dT}^*$  and  $N_{dCCN}^*$ . Section 5.2.3 analyzes the agreement between  $N_d^*$  and  $N_a$ .

### 5.2.1 Comparison between $N_d$ measurements with estimated $N_{dT}$ and $N_{dCCN}$

The PMM procedure was applied to the measured  $W_b$  and  $N_d$  for analyzing the spectrum of  $N_d$ ,  $N_{dT}$  and  $N_{dCCN}$  values near cloud base (as described in Section 4.2). This analytical method makes it possible to identify the role of  $W_b$  in producing  $N_d$ . A perfect agreement of the values is not expected due to the turbulent nature of the clouds, but the statistical modes of the measurements should have similar values to the theoretical estimation of the same modes of  $N_{dCCN}$  and  $N_{dT}$ , within their uncertainty range. Figures 11 and 12 show  $N_{dCCN}$ ,  $N_{dT}$ , and  $N_d$  values for the two cloud

540 probes as a function of  $W_b$  for the cases presented in Table 3. The uncertainties regarding the  $S_{max}$ ,  $N_{dCCN}$  and  $N_{dT}$  estimates for measurements at cloud base with both probes (CCP-CDP and CAS-DPOL) are on average about 22, 20, and 38 % for all flights, respectively (the uncertainty methods adopted for these theoretical estimates are available in Appendix A). The  $W_b$  uncertainty of  $0.3 \text{ m s}^{-1}$  accounts on average for about 65% (60 %) of the  $N_{dT}$  ( $S_{max}$ ) uncertainty, and the uncertainty from the estimated  $S_{max}$  contributes most of the  $N_{dCCN}$  uncertainty (~70% on average).  
545

Both values of  $N_{cas}$  and  $N_{cdp}$  are within the range of the theoretical expectation of  $N_{dT}$  and  $N_{dCCN}$ , except for occasional deviations at the extreme percentiles. For example, the maximum  $N_{dT}$  versus maximum  $N_d$  are outside the error interval for  $N_{dT}$ . This is so because extreme percentiles are much more prone to random variations than the middle range, such as the median.

550 The curves for  $N_{dT}$  mostly agreed quite well with those for  $N_d$  with only small deviations. The  $N_{dCCN}$  mostly underestimates  $N_d$  by as much as a factor of 0.5 for reasons that we could not identify. Entrainment is not a likely cause, because it would dilute  $N_d$  and thus incur  $N_{dCCN}$  to be biased positively with respect to  $N_d$ . It appears that measuring  $S$  in clouds is still a great challenge, even indirectly by using Eq. 3. Remarkably, Eq. 2 (Twomey, 1959), which avoids an explicit usage of  $S$ , still performs better when limited within the observed bounds of  $W_b$  and  $S$  within the cloud. These results support the analyses concerning the  $N_d$  measurement at cloud base that are presented  
555 in the next sections.

### 5.2.2 Comparing estimated $N_d^*$ with $N_{dT}^*$ and $N_{dCCN}^*$

Assuming that  $W_b^*$  represents the updraft velocity for a set of cloud base measurements, the corresponding  
560 measured  $N_d^*$  from CAS-DPOL and CCP-CDP ideally should have similar values to the estimated  $N_{dCCN}^*$  and  $N_{dT}^*$ . The uncertainties of  $N_{dCCN}^*$  and  $N_{dT}^*$  are ~20 % and ~35 % on average, respectively. Figure 13a shows the values of  $N_d^*$  and  $N_{dT}^*$  for the different cloud base measurements shown in Figs. 11 and 12. The  $N_{dT}^*$  agrees with  $N_d^*$  within the measurements uncertainties, as shown by the error bars. The bias of  $N_{dT}^*$  with respect to  $N_d^*$  for the CAS -DPOL is 1.00 with a standard deviation  $\pm 0.17$  around it. The respective result for the CDP is  $0.84 \pm 0.12$ . A weaker agreement is observed for comparisons between  $N_{dCCN}^*$  and  $N_d^*$  (see Fig. 13b). A factor of ~2 can be observed for  
565 some cases (AC14 and AC17). The bias of  $N_{dCCN}^*$  with respect to  $N_d^*$  for the CAS-DPOL is  $0.80 \pm 0.07$ . The respective result for the CDP is  $0.76 \pm 0.1$ .

### 5.2.3 Comparing estimated $N_d^*$ with $N_a$

570 Another possibility of cloud base closure is via comparison of  $N_d^*$  and  $N_a$  estimates from measurements of the vertical evolution of  $r_e$  in pristine and polluted conditions. In these situations, the estimated values for these parameters is expected to converge. Figure 14a shows the calculated  $N_a$  with CCP-CDP probe results from cloud measurements during flight AC17. The estimated  $N_a$  in this case is  $1496 \text{ cm}^{-3}$ , and, considering evaporation losses due to cloud mixing, the expected number of droplets at cloud base is  $1047 \text{ cm}^{-3}$  after applying the correction by a  
575 reduction of 30% (Freud et al., 2011), which is a factor of 0.7.  $N_d^*$  for the same flight segment is  $1207 \text{ cm}^{-3}$ , calculated from CCP-CDP data (see Fig. 7b). The factor of 0.7 applied to the estimates of  $N_a$  corroborates the



methodology of Freud et al. (2011) for retrieving the effective number of droplets nucleated at cloud base, even though different dataset was used in here. A close relationship between  $M_v$  and  $r_e$  as a function of height is shown at Figure 14b. Similar results were found for cloud profile measurements during the other flights.

580 Values of  $N_a$  and  $N_d^*$  were calculated for all profile flights and cloud probes, and the results are shown in Fig. 15. The uncertainties of  $N_a$  ( $N_d^*$ ) estimates from CAS-DPOL and CCP-CDP are ~25% (21 %) and ~14% (10%), respectively. The comparisons between the estimated  $N_a$  and  $N_d^*$  show a linear correlation with correlation coefficients greater than 0.97 for all cloud probes. The linear regression between  $N_d^*$  and  $N_a$  estimates shows a slope close to one for CAS-DPOL and CCP-CDP. More specifically, the bias of  $N_a$  with respect to  $N_d^*$  for the CAS -  
585 DPOL is  $1.12 \pm 0.19$ . The respective result for the CDP is  $0.78 \pm 0.19$ .

The lower slope for the CDP (Fig. 15b) than for the CAS-DPOL (Fig. 15a) can be explained by the relative overestimate of  $r_e$  of the CDP compared to the CAS-DPOL, which translates to an underestimate in  $N_a$ . This is inferred from Fig. 4, which shows that the CDP has about 20% more CWC compared to the CAS-DPOL and the hot wire CWC.

590 These results show good agreement with theoretical expectations, especially when based on the CAS-DPOL. The flights performed in near-pristine and polluted conditions can be distinguished based on the CAS-DPOL estimates of  $N_d^*$  and  $N_a$  values. For example, in flight AC19 performed over the Atlantic Ocean in clean conditions, the CAS-DPOL estimated values of  $N_d^*$  and  $N_a$  are  $\sim 270 \text{ cm}^{-3}$ , whereas for flights AC07 and AC11 performed under polluted conditions, the values of  $N_d^*$  and  $N_a$  are greater than  $1000 \text{ cm}^{-3}$ .

595 In general, the similarities of  $N_a$  and  $N_d^*$  values (see Figure 15) supports the methodology to calculate the effective number of droplets observed at cloud base of convective clouds from the vertical profile of measured  $r_e$  or  $M_v$ . Some qualification might exist for the most polluted flights, e.g., AC08, where  $N_a$  is larger than  $N_d^*$  by a factor of ~20-30%. The vertical profiles of the  $N_d$  measurements indicate that in these cases the  $N_d$  measurements up to 2-3 kilometers above cloud base were larger than those at cloud base. A higher aerosol concentration at these greater  
600 heights was also observed in aerosol probe measurements (not shown), suggesting that secondary droplet nucleation was taking place on the most polluted flights. The  $N_a$  calculation does not take into account the possibility of new nucleation above cloud base (Freud et al., 2011). Therefore, the assumption of adiabatic growth of droplets via condensation from cloud base to higher levels within cloud can lead to an overestimation by ~20-30% of the number of droplets at cloud base when calculating  $N_a$  in cases with secondary droplet nucleation.

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## 6. Summary and conclusions

This study is focused on testing recent parameterizations that are used for the recently developed methodology of satellite retrievals of  $N_a$ ,  $W_b^*$ , and CCN in convective clouds, based on aircraft measurements during the ACRIDICON-CHUVA campaign in the Amazon. It is the first time that these new parameterizations are tested com-  
610 prehensively alongside old parameterizations. Liquid water content measurements from a hot-wire device were taken as a reference for the quality assessment of estimated CWC from cloud probe DSDs near cloud base. The intercomparison of the DSDs and the CWC derived from the different instruments generally shows good agreement within the instrumental uncertainties. The values of  $N_d$  near cloud base were comparable within the measurement

615 errors with their inferred values based on the measured  $W_b^*$  and  $N_{CCN}(S)$ . The values of  $W_b^*$  were calculated from  
the measured spectrum of  $W_b$  using the parameterization of Rosenfeld et al. (2014a), which is also used for retriev-  
ing cloud base updraft from satellites (Zheng et al., 2015). In addition,  $N_d$  near cloud base compared well with  $N_a$   
(within  $\pm 20\%$ ), obtained from the vertical evolution of cloud drop effective radius ( $r_e$ ) above cloud base. The values  
of  $N_a$  in this study were obtained with the same parameterization that has been recently developed for satellite calcu-  
620 lated  $N_a$  based on the satellite retrieved vertical evolution of  $r_e$  in convective clouds (Freud et al., 2011; Rosenfeld et  
al., 2014a). These results support the methodology to derive  $N_a$  based on the rate of  $r_e$  growth with cloud depth and  
under the assumption that the entrainment and mixing of air into convective clouds is extremely inhomogeneous.  
The measured effective droplet numbers ( $N_d^*$ ) at cloud base were also compared against  $N_{dT}^*$  which is its predicted  
value based on the old parameterization in Eq. 2 (Twomey, 1959), which uses  $W_b^*$  and the  $N_{CCN}(S)$  power law. A  
newer parameterization calculates  $N_{dCCN}^*$  by substituting  $S$  into the power law  $N_{CCN}(S)$ , where  $S$  is obtained from  
625 Eq. 3 (Pinsky et al., 2012). The agreement between  $N_d^*$  and  $N_{dCCN}^*$  was only within a factor of 2, underlying the yet  
unresolved challenge of aircraft measurements of  $S$  in clouds.  
In summary, the measurements of  $N_{CCN}(S)$  and  $W_b$  did reproduce the observed  $N_d$  when using Twomey's parameteri-  
zation, while using measured in-cloud  $S$  remains a challenge. Furthermore, the vertical evolution of  $r_e$  with height  
reproduced the observation-based adiabatic cloud base drop concentrations,  $N_a$ . The combination of these results  
630 provide aircraft observational support for the various components of the satellite retrieval methodology that was  
recently developed to retrieve  $N_{CCN}(S)$  below the base of convective clouds (Rosenfeld et al., 2016).

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## Appendix – A

The uncertainties of  $N_{dT}$ ,  $N_{dCCN}$ ,  $S_{max}$ ,  $r_e$ ,  $CWC$ ,  $N_a$ ,  $N_d^*$ ,  $N_{dT}^*$  and  $N_{dCCN}^*$  were estimated via the ‘analytic’ approxima-  
tion of error propagation which is calculated by finding the partial derivatives of the function with respect to the  
920 independent variables. For example if you have a function  $f(x,y,z)$  with the variables  $x,y$  and  $z$  and the uncertainties  
 $s_x$ ,  $s_y$  and  $s_z$  you have the following result for the final uncertainty ( $sf$ ) of  $f$ :

$$sf = \sqrt{\left(\frac{\delta f}{\delta x}\right)^2 \cdot s_x^2 + \left(\frac{\delta f}{\delta y}\right)^2 \cdot s_y^2 + \left(\frac{\delta f}{\delta z}\right)^2 \cdot s_z^2} \quad (A1)$$

## Appendix - B

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### Calculating STDE CCNmax and STDE CCNmin

The  $N_0$  and  $k$  parameters standard errors (STDE) are associated with the statistical uncertainty of the power law function fit. To compute the STDE for the CCN estimates the uncertainties of  $S$  (~10%) are considered. Then, the maximum and the minimum STDE values expected for the CCN estimates are calculated as follows:

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*Maximum STDE*

$$STDE\ CCN_{max} = \frac{[(N_0 + SD \cdot N_0) \cdot (S_i \cdot 1.1)^{k+SD \cdot k}] - N_0 \cdot S_i^k}{\sqrt{N}} \quad (A2)$$

where:

The averaging is done on  $I=1:N$ .

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$N$  is the number of  $NCCN_2$  cases for each group of measurements.

$SD \cdot N_0$  is the statistical standard deviation of  $N_0$ ;

$SD \cdot k$  is the statistical standard deviation of  $k$ ;

$S_i$  is the supersaturation in each step, forced to have the maximum value (multiplied by 1.1).

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*Minimum STDE*

$$STDE\ CCN_{min} = \frac{N_0 \cdot S_i^k - [(N_0 - SD \cdot N_0) \cdot (S_i \cdot 0.9)^{k-SD \cdot k}]}{\sqrt{N}} \quad (A3)$$

where:

The averaging is done on  $I=1:N$ .

$N$  is the number of  $NCCN_2$  cases for each group of measurements.

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$SD \cdot N_0$  is the statistical standard deviation of  $N_0$ ;

$SD \cdot k$  is the statistical standard deviation of  $k$ ;

$S_i$  is the supersaturation in each step, forced to have the minimum value (multiplied by 0.9).

### Error and uncertainties of Twomey's formula (Eq. 1)

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According to Krüger et al. (2014), the error in  $N_{CCN}(S)$  based on the counting error of the measured particle number ( $\Delta c = \sqrt{c + 1}$ ) and can be calculated by:

$$\Delta N = \sqrt{\left(\frac{\Delta c \cdot t}{q}\right)^2 + \left(\frac{c \cdot \Delta q \cdot t}{q^2}\right)^2 + \left(\frac{c \cdot \Delta t}{q}\right)^2} \quad (A4)$$

where:

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$t$  is the period of the time of measurements assumed (60 s);

$\Delta t$  is the error of the time;



c is the measured particle number;

q is the aerosol flow rate;

$\Delta q$  is the error of the aerosol flow rate (we assume 10% of  $q$ , i.e.  $0.007 \text{ L min}^{-1}$ );

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According to the Gaussian error propagation the error in  $N_{CCN}(S)$  is:

$$\Delta N_{CCN} = \sqrt{\left(\frac{\Delta m_{CCN2} * T_{m_{CCN1}}}{m_{CCN1}}\right)^2 + \left(-\frac{m_{CCN2} * \Delta m_{CCN1}}{\frac{m_{CCN1}^2}{T_{m_{CCN1}}}}\right)^2 + \left(\frac{\Delta T_{m_{CCN1}} * T_{m_{CCN2}}}{m_{CCN1}}\right)^2} \quad (A5)$$

To calculate the error in  $k$  the calculations have been done with the upper and lower error ranges and the resulting error of  $k$  is 20 % of  $k$ . For  $N_0$  the calculated error is 23 %, associated with the Twomey equation fit (Eq. 1) and the  $N_{CCN}$  error.

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### Appendix – C

Summary of the measurements and theoretical calculations at cloud base:

- 1)  $N_d$  - based on probe measurement;
- 2)  $N_a$  - based on vertical profile of  $r_e$ ;
- 3)  $S_{max} - S$  substituting  $N_d$  and  $W_b$  in Eq. 3.
- 4)  $N_{dT}$  - Obtained from substituting in Eq. 2  $W_b$  and  $N_{CCN}(S)$  parameters ( $k$  and  $N_0$ );
- 5)  $N_{dCCN}$  - Obtained from substituting  $S_{max}$  and  $N_{CCN}(S)$  parameters in Eq. 1.
- 6)  $W_b^*$  - Obtained from Eq. 6.
- 7)  $N_d^*$ ,  $N_{dT}^*$ ,  $N_{dCCN}^*$  -  $N_{db}$ ,  $N_{dT}$ ,  $N_{dCCN}$  that match  $W_b^*$ .

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### Figure captions

Figure 1. Flight patterns below and in convective clouds during the ACRIDICON-CHUVA campaign.

Figure 2. HALO flight tracks during the ACRIDICON-CHUVA experiment. The flight numbers are indicated on the right (from Wendisch et al., 2016).

980

Figure 3. Cloud droplet effective radius ( $r_e$ ) as a function of altitude for clouds over clean (Flight AC19 - blue squares), polluted (Flight AC18 – green triangles) and very polluted (Flight AC13 – brown diamonds) environments. Dashed lines indicate the probability of rain from the coalescence process expressed as percentage on the top of the graphic.

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Figure 4. Mean cloud water content from the hot-wire measurements and estimated from the cloud probes (CCP-CDP and CAS-DPOL from top to bottom, respectively) as a function of effective radius ( $r_e$ ) size (left panel). The ratios between the hot-wire liquid water content and the cloud water content derived from each probe are shown in red ( $CWC_r$ ). The total uncertainties for each probe and the hot-wire measurements are shown by the dotted lines.

The number of cases (black continuous line), hot-wire measurement standard deviations (dashed black line), and probe CWC standard deviations (dashed colored line) for each  $r_e$  size are shown in the right panels.

990 Figure 5. Mean cloud droplet concentrations for CAS-DPOL and CCP-CDP as a function of effective radius ( $r_e$ ) (left panel). The systematic error for each probe is shown by the dashed line. The right panel indicates the standard deviation in  $\text{cm}^{-3}$  of each probe concentration as a function of  $r_e$ . The probes are identified by colors as shown at the top of the panels. The sample for each probe is the same as shown in Figure 4.

995 Figure 6. Mean cloud droplet concentration (solid lines) and cloud water content as a function of droplet diameter in the left and right panels, respectively, for a)  $5 \mu\text{m} < r_e < 6 \mu\text{m}$ ; b)  $8 \mu\text{m} < r_e < 9 \mu\text{m}$ ; c)  $11 \mu\text{m} < r_e < 12 \mu\text{m}$ ; d)  $12 \mu\text{m} < r_e < 13 \mu\text{m}$ . The probes are identified by colors as shown at the top of the panels. The error bars indicate the uncertainty range of mean cloud droplet concentration and cloud water content values as a function of droplet diameter.

1000 Figure 7. a) Frequency histogram of vertical wind speed ( $W_b$ ) from cloud base measurements on flight AC17 (labeled on the left ordinate). The blue line indicates the cumulative probability function of  $W_b$  (labeled on the right ordinate). The cyan arrow indicates the value of  $W_b^*$  ( $1.83 \text{ m s}^{-1}$ ) that represents the 86th percentile of the  $W$  spectra; b) Similar for the cloud droplet concentrations measured with the CCP-CDP probe. The cyan line indicates the  $N_d^*$  value ( $1207 \text{ cm}^{-3}$ ) at the 86th percentile in the  $N_d$  spectra. The indicated time is in UTC and shows the time of the first cloud penetration at cloud base and the total number of 1-s measured cloud data points.

1005 Figure 8.  $CCN_1$  (red dots) and  $CCN_2$  (black dots) measurements for a segment of flight AC17 on 27 September 2014. The abscissa shows the measurement time in UTC. The blue line indicates the altitude in meters above sea level and is labeled on the left ordinate (as well as  $CCN_1$  and  $CCN_2$ ).  $S_1$  and  $S_2$  measurements in % are indicated by the orange and green lines, respectively (both are labeled on the right ordinate). Cyan dots on the blue line indicate cloud penetrations (i.e., when cloud droplet concentrations are greater than  $20 \text{ cm}^{-3}$ ). In this case, cloud base heights were observed around 2,300 meters above ground.

1010 Figure 9. A comparison of the CCN spectra derived from the two CCN counter columns on board the HALO aircraft during flight AC17. Black (blue) smaller dots indicate  $CCN_1$  ( $CCN_2$ ) measurements for each second. Large diamonds in black (blue) indicate the  $mCCN_1$  ( $mCCN_2$ ) for each time step of measurements. The orange large diamonds indicate the  $NCCN_2$  values that are used to fit the power law equation of the group of measurements, which is shown at the lower right corner of the plot. The standard error for the CCN spectra derived is shown in Table 2.

1015 Figure 10. CCN spectra as measured on board the HALO aircraft during cloud profiling flights. Diamonds indicate the  $NCCN_2$  values, which are used to fit the power law equation of the group of measurements. The colors indicate the group of measurements and match the legend on the right side of the plot. The legend indicates the flight number; the initial time of group measurements; the period of measurements in seconds; the power law fit and the correlation coefficient of the data. The standard errors for each CCN spectra derived are shown in Table 2.

1020 Figure 11a-f.  $N_{dCCN}$ ,  $S$ ,  $N_{dT}$  and  $N_d$  values are presented as a function of the cloud base updrafts ( $W_b$ ). This plot is based on the ‘probability matching method’ (PMM), using same percentiles for  $W_b$  and  $N_d$  ( $N_{dCCN}$  or  $N_{dT}$ ). The values of  $N_{dCCN}$ ,  $N_{dT}$  and  $N_d$  are shown the left y-axis, those of  $S$  on the right y-axis. The black dashed lines are the

1025  $N_{dT}$  uncertainties. The gray solid (dashed) lines are the  $N_{dCCN}$  values (uncertainties). The effective updraft  $W_b^*$  for each flight segment is shown by the cyan line. The data are based on the CAS-DPOL probe. The time, period of measurements (sample size in seconds), and  $N_{CCN}(S)$  equation are shown on the top of the figures.

Figure 12a-d. Same as Figure 11 for the CCP-CDP probe. No data were available for flight AC16. The CCP-CDP malfunctioned in flight AC13 during the cloud base measurements.

1030 Figure 13. a)  $N_d^*$  versus  $N_{dT}^*$  calculated with  $W_b^*$  from cloud base data shown in Figures 11-12. The CAS-DPOL values are indicated by plus symbols (+) and the CCP-CDP values are indicated by circles (o). The colors indicate each flight segment (legend in the right side of the plot). Error bars indicate the uncertainties of variable estimates. Lines show the 1:1 and 1:2 relationships between  $N_{dT}^*$  versus  $N_d^*$  for each probe; b) Same for  $N_d^*$  versus  $N_{dCCN}^*$ .

1035 Figure 14 a). Mean volume drop mass ( $M_v$ ) versus liquid water content from the CCP-CDP measurements for adiabatic fraction greater than 0.25 ( $LWC_a$ ). Values are shown with different colors labeled as a function of height in kilometers above sea level (indicated by the colorbar on the right side of the graphic). The slope of the linear equation is the estimated  $N_a$  (i.e.,  $1496 \text{ cm}^{-3}$ ); b)  $M_v$  versus  $r_e$  as a function of height in kilometers above sea level (indicated by the color bar on the right side of the graphic).

1040 Figure 15.  $N_d^*$  versus  $N_a$  measured with CAS-DPOL and CCP-CDP (indicated on the top of panels) for profile flights during the ACRIDICON-CHUVA campaign. The color of the dots is associated with the flight number shown at the right side of the panels. Error bars indicates the uncertainties of variables estimates. The linear regression equation and the correlation coefficient  $R$  are shown at the top of each panel.

#### 1045 **Table captions**

Table 1. Cloud probe size intervals and central bin diameters during HALO flights.

1050 Table 2. Estimates of  $N_0$  and  $k$  below cloud base and their standard error (STDE) for each case study. Maximum and minimum STDE (STDE  $CCN_{max}$  and STDE  $CCN_{min}$ , respectively) for the  $CCN$  measurements are calculated considering errors in the supersaturation measurements ( $\sim 10\%$ ). The details about the calculation of these values are given in Appendix B.

Table 3. List of case studies for measurements below cloud base. The duration of measurements is given in seconds, starting at the initial time indicated. An asterisk indicates those flights where the two probes provided at least 20 seconds of measurements at cloud base. The data can be from different cloud passes in the same region of measurements below cloud base.

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**Tables**

Table 1. Cloud probe size intervals and central bin diameters during HALO flights.

<b>Cloud Probe</b>	<b>Size interval</b>	<b>Number of bins</b>	<b>Central bin diameter (<math>\mu\text{m}</math>)</b>
<b>CCP-CDP</b>	3-50 $\mu\text{m}$	14	3.8, 6.1, 8.7, 10.9, 13.5, 17.1, 19.7, 22.5, 25.9, 28.3, 31.7, 36.6, 40.7, 44.2
<b>CAS-DPOL</b>	3-50 $\mu\text{m}$	10	3.9, 6, 10.8, 17.3, 22.3, 27.4, 32.4, 37.4, 42.4, 47.4

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Table 2. Estimates of  $N_0$  and  $k$  below cloud base and their standard error (STDE) for each case study. Maximum and minimum STDE (STDE  $CCN_{max}$  and STDE  $CCN_{min}$ , respectively) for the  $CCN$  measurements are calculated considering errors in the supersaturation measurements ( $\sim 10\%$ ). The details about the calculation of these values are given in Appendix B.

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Flight	Time	$N_0$	$k$	STDE $N_0$	STDE $k$	STDE $CCN_{max}$ [ $\text{cm}^{-3}$ ]	STDE $CCN_{min}$ [ $\text{cm}^{-3}$ ]
AC11	14:58:21	1985	0.73	81.6	0.035	25.5	24.8
AC11	17:38:20	2927	1.14	82.8	0.032	43.9	43.8
AC12	15:56:00	1764	0.3	71.4	0.046	19.0	22.7
AC13	16:29:01	4145	0.92	64.7	0.016	69.7	54.8
AC14	15:21:40	1509	0.97	44.8	0.028	24.7	18.9
AC15	13:33:35	2209	0.94	70.4	0.038	47.4	31.2
AC16	20:21:40	1966	0.67	69.5	0.029	26.5	21.2
AC17	16:50:50	2743	0.72	38.7	0.013	31.9	30.5
AC17	19:38:20	1015	0.54	18.5	0.018	10.7	9.4

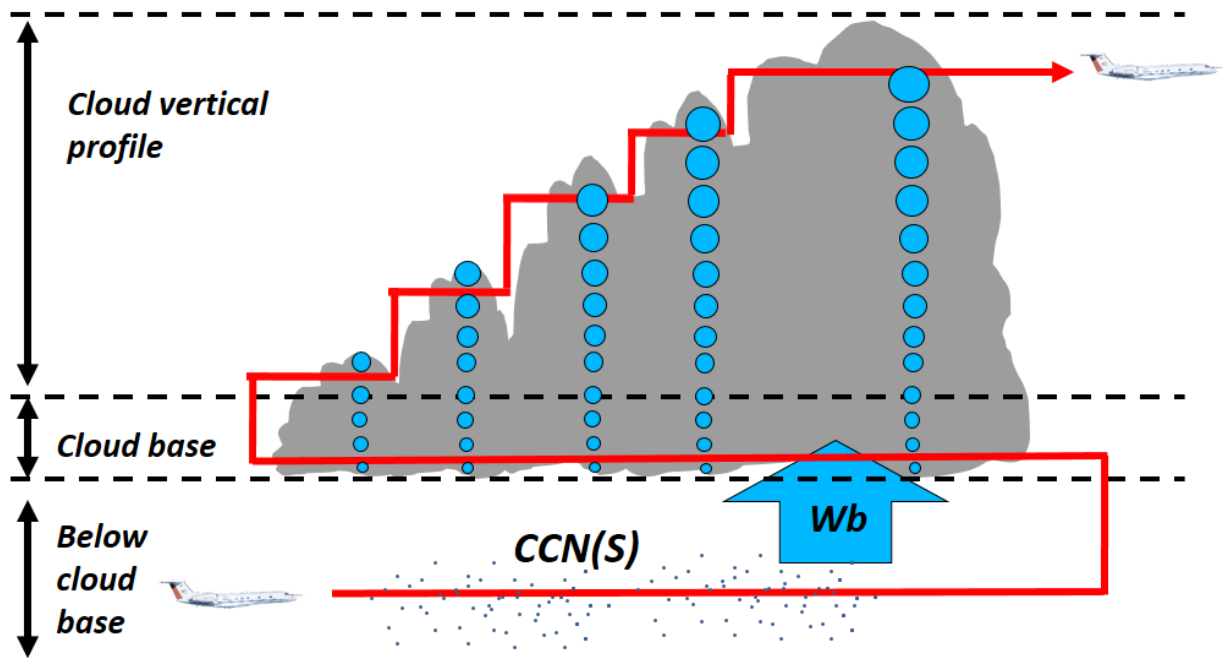
Table 3. List of case studies for measurements below cloud base. The duration of measurements is given in seconds, starting at the initial time indicated. An asterisk indicates those flights where the two probes provided at least 20 seconds of measurements at cloud base. The data can be from different cloud passes in the same region of measurements below cloud base.

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<b>Measurements below cloud base</b>			
<b>Flight</b>	<b>Date</b>	<b>Initial time (UTC)</b>	<b>Period of analysis (s)</b>
AC11	16/09/2014	14:58:21	593
AC11*	16/09/2014	17:38:20	710
AC12	18/09/2014	15:56:00	440
AC13*	19/09/2014	16:29:01	722
AC14*	21/09/2014	15:21:40	800
AC15	23/09/2014	13:33:35	555
AC16*	25/09/2014	20:21:40	550
AC17*	27/09/2014	16:50:50	831
AC17*	27/09/2014	19:38:20	840

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Figures



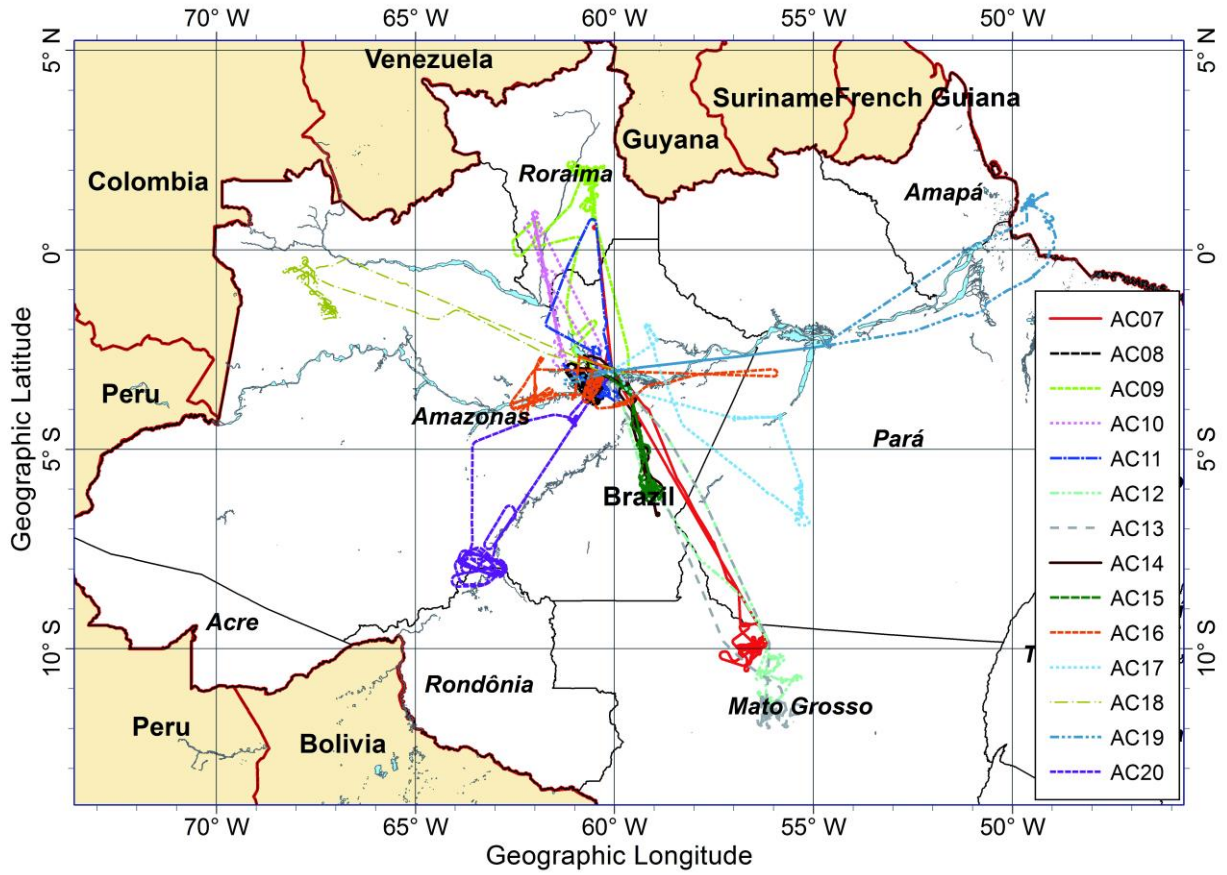
1080 Figure 1. Flight patterns below and in convective clouds during the ACRIDICON-CHUVA campaign.

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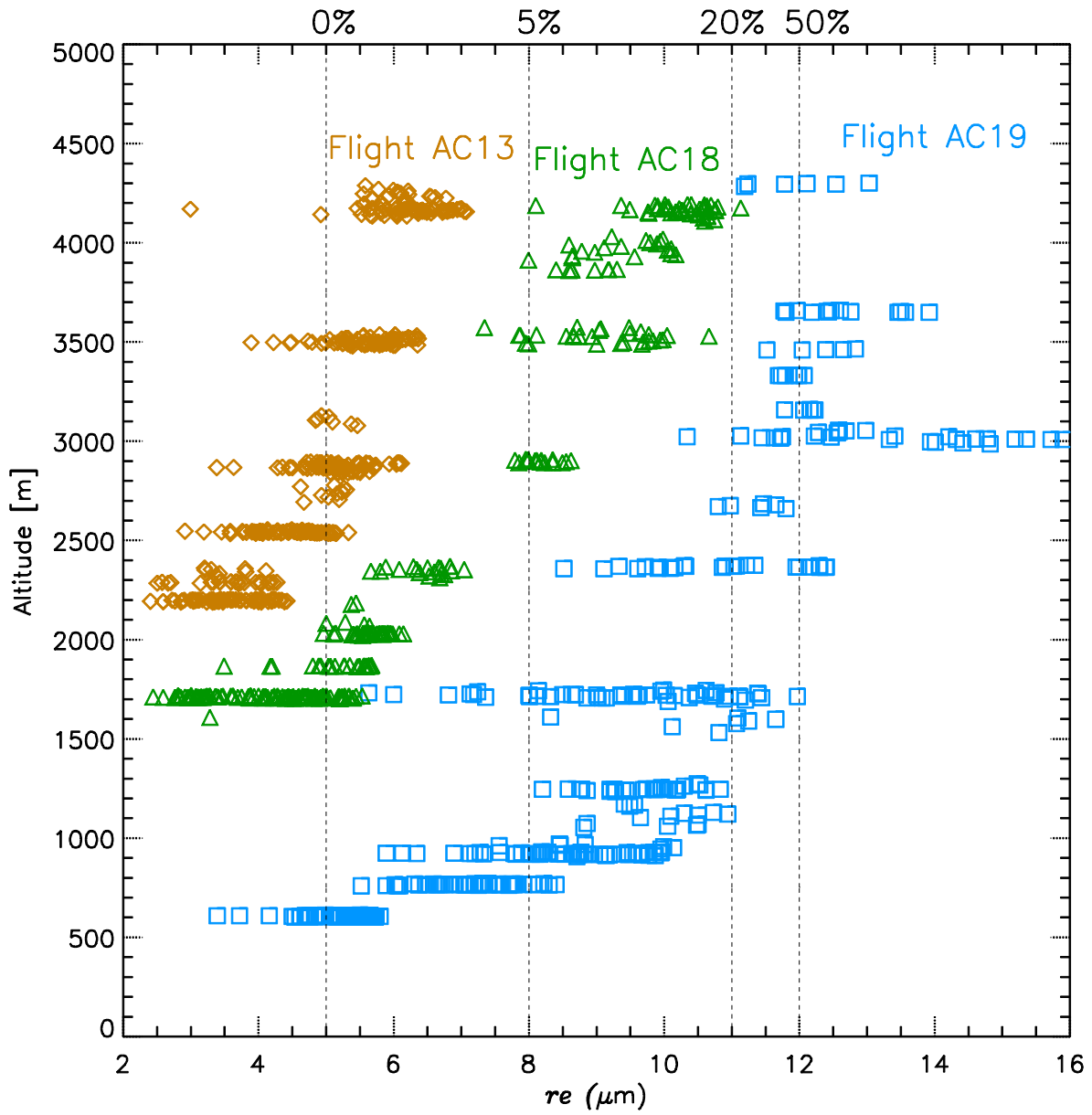
Figure 2. HALO flight tracks during the ACRIDICON-CHUVA experiment. The flight numbers are indicated on the right (from Wendisch et al., 2016).

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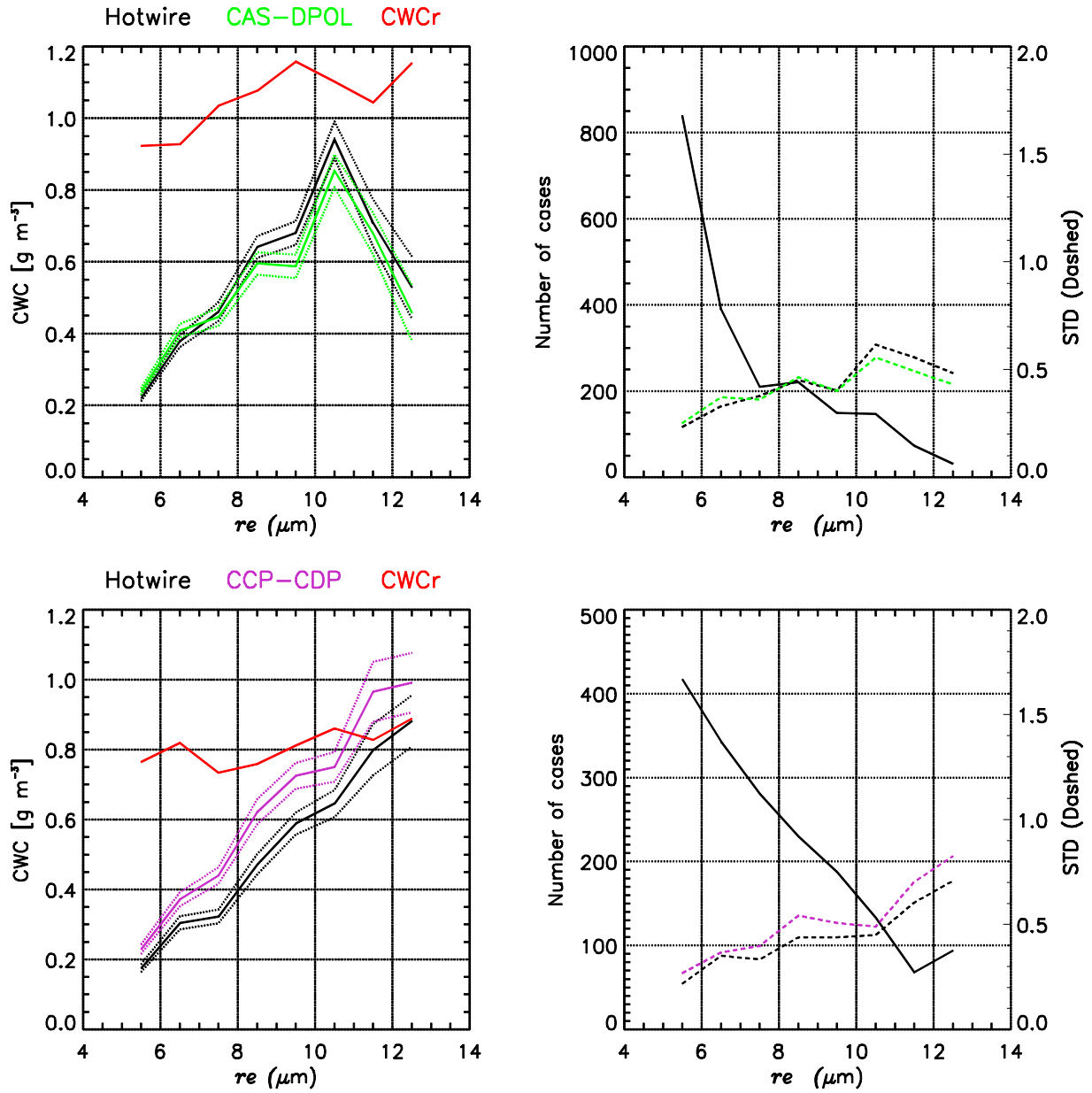
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Figure 3. Cloud droplet effective radius ( $r_e$ ) as a function of altitude for clouds over clean (Flight AC19 - blue color squares), polluted (Flight AC18 - green color triangles) and very polluted (Flight AC13 - brown color diamonds) environments. Dashed lines indicate the probability of rain from the coalescence process expressed in percentage on the top of the graphic.

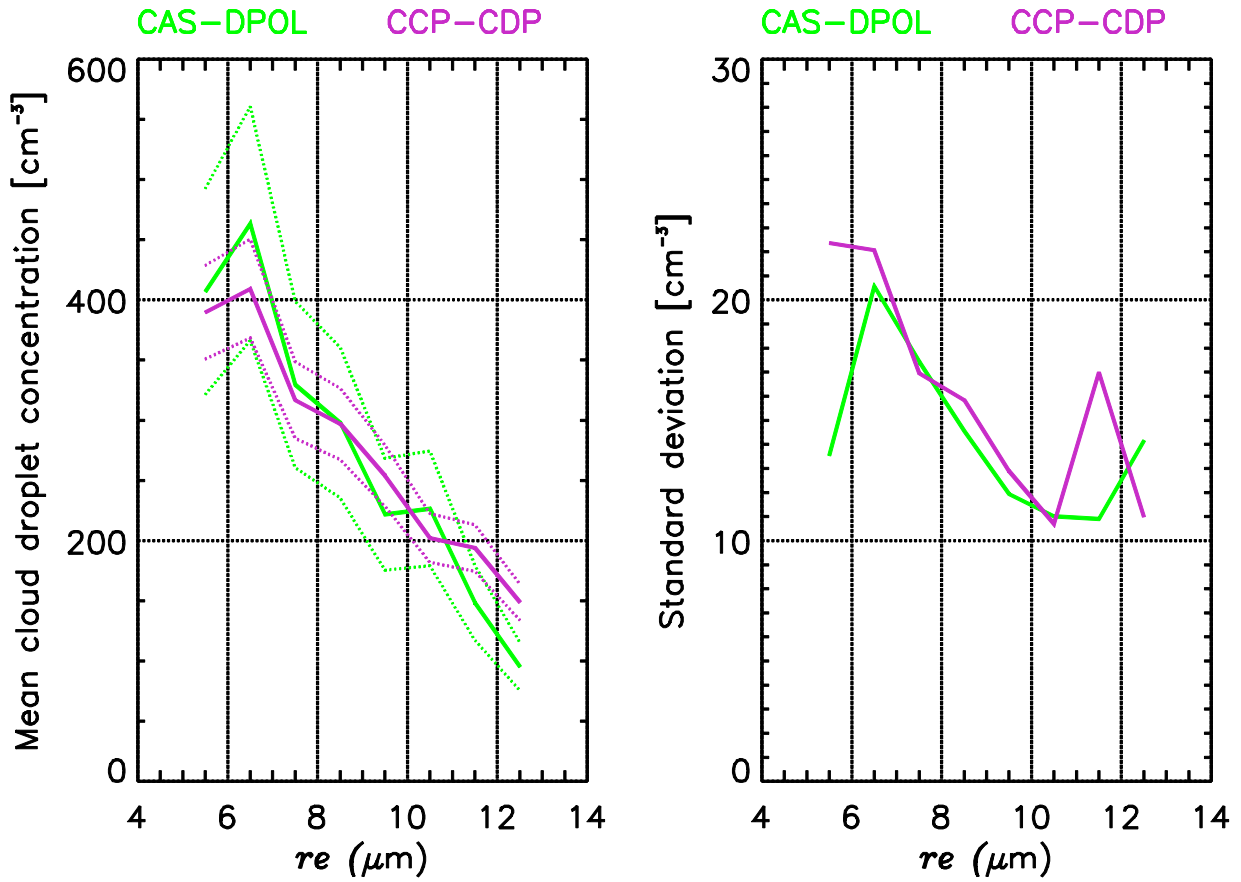




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Figure 4. Mean cloud water content from the hot-wire measurements and estimated from the cloud probes (CCP-CDP and CAS-DPOL from top to bottom, respectively) as a function of effective radius ( $r_e$ ) size (left panel). The ratios between the hot-wire liquid water content and the cloud water content derived from each probe are shown in red ( $CWC_r$ ). The total uncertainty for each probe and the hot-wire measurements are shown by the dotted lines. The number of cases (black continuous line), hot-wire measurement standard deviations (dashed black line), and probe CWC standard deviations (dashed colored line) for each  $r_e$  size are shown in the right panels.

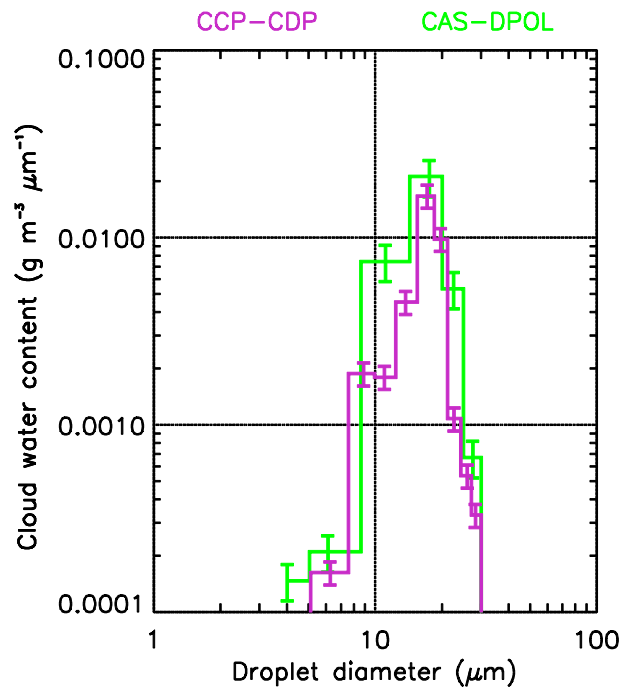
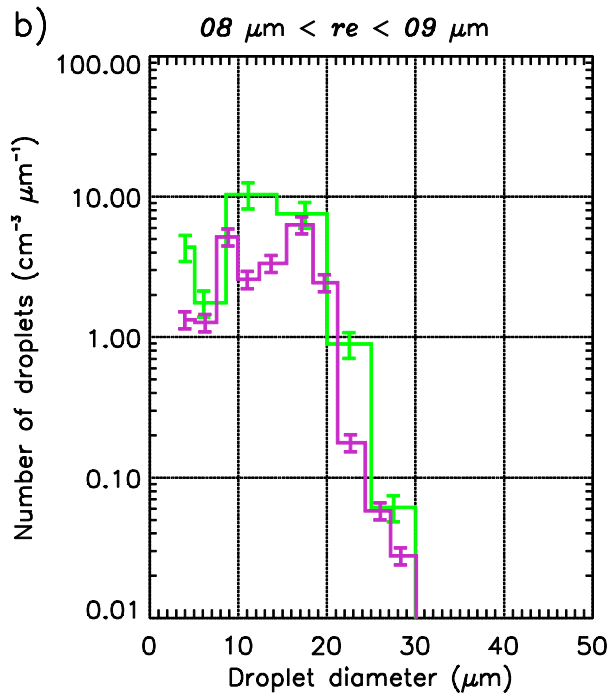
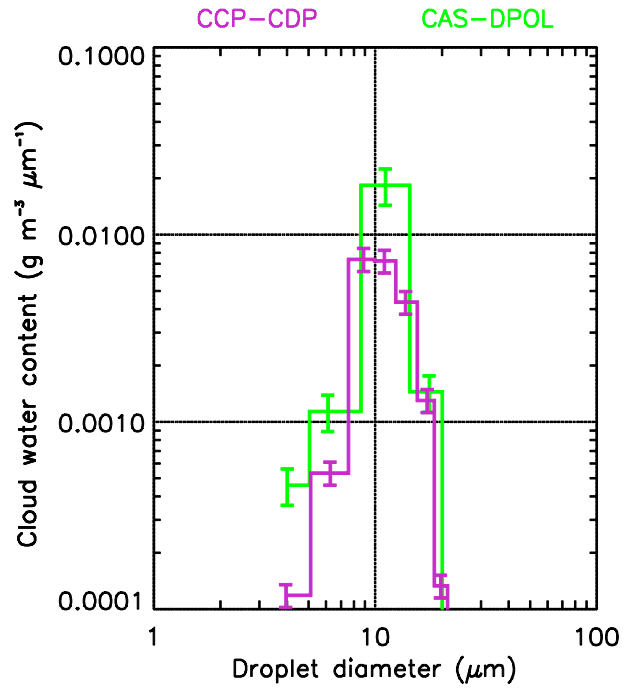
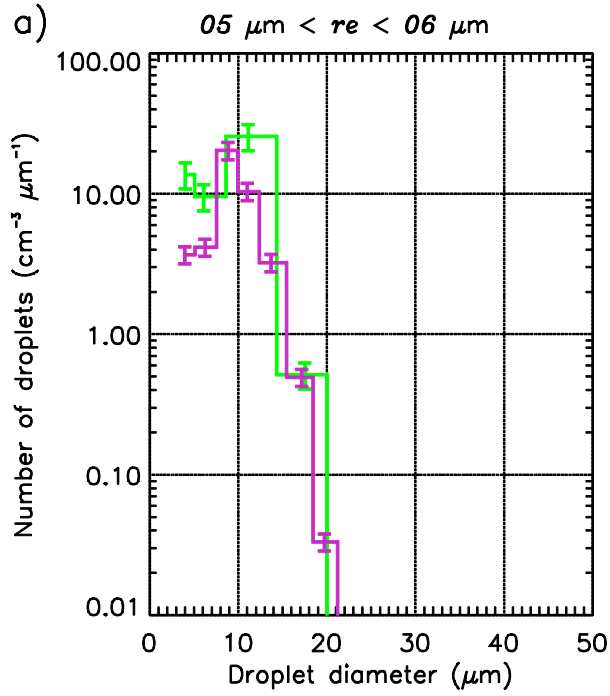


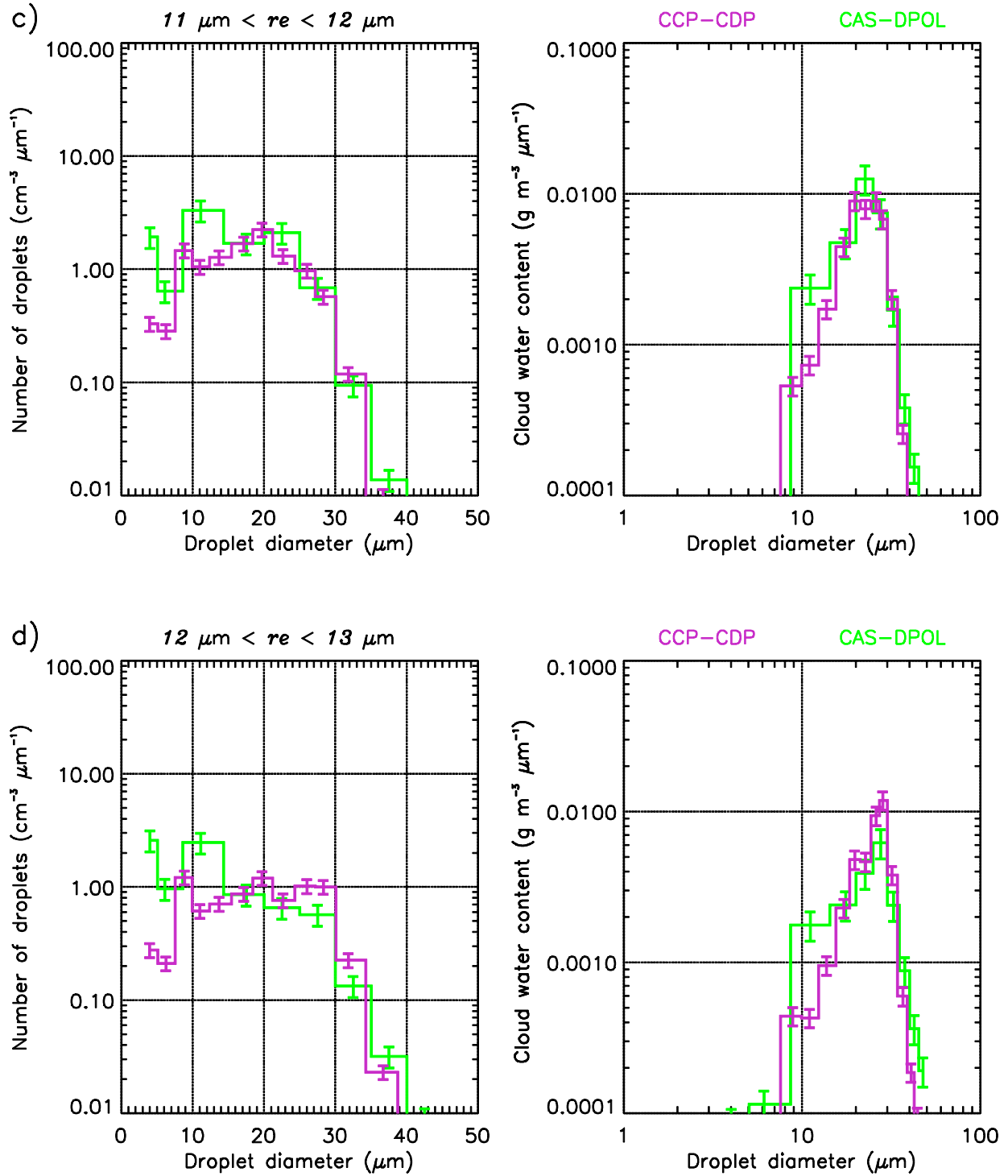
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Figure 6. Mean cloud droplet concentration (solid lines) and cloud water content as a function of droplet diameter in the left and right panels, respectively, for a)  $5 \mu\text{m} < r_e < 6 \mu\text{m}$ ; b)  $8 \mu\text{m} < r_e < 9 \mu\text{m}$ ; c)  $11 \mu\text{m} < r_e < 12 \mu\text{m}$ ; d)  $12 \mu\text{m} < r_e < 13 \mu\text{m}$ . The probes are identified by colors as shown at the top of the panels. The error bars indicate the uncertainty range of mean cloud droplet concentration and cloud water content values as a function of droplet diameter.

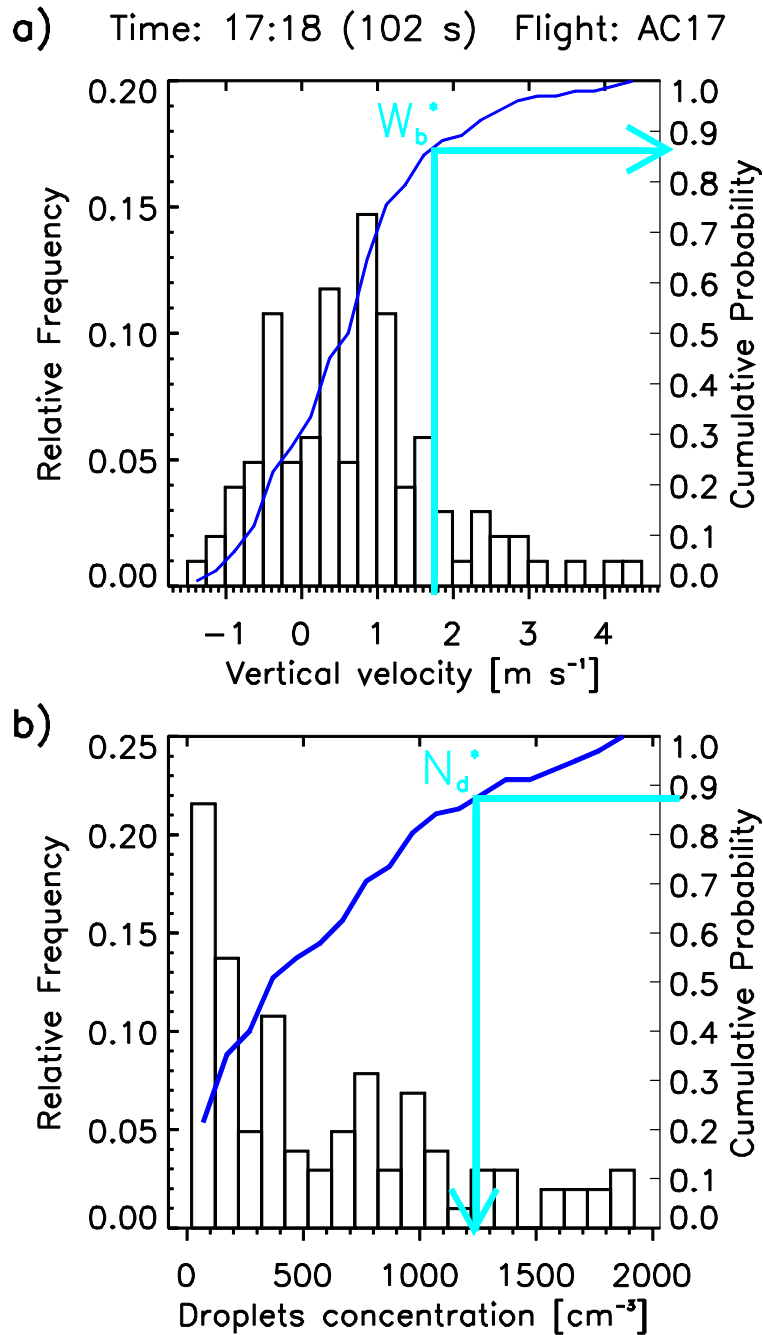
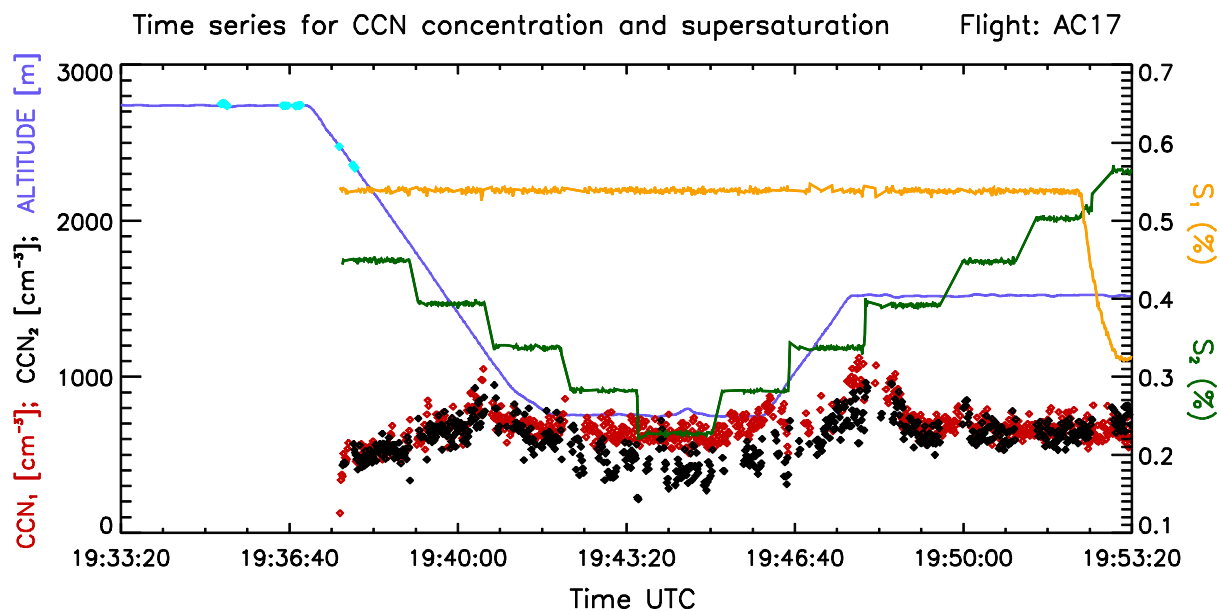


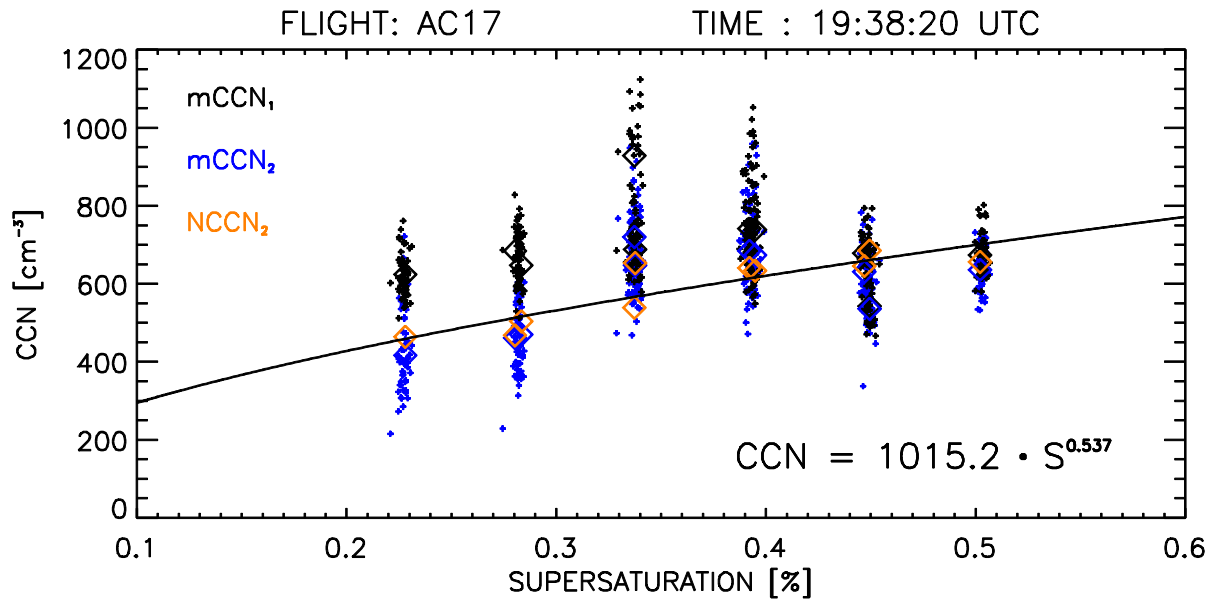
Figure 7. a) Frequency histogram for vertical wind speed ( $W_b$ ) from cloud base measurements on flight AC17 (labeled on the left ordinate). The blue line indicates the cumulative probability function of  $W_b$  (labeled on the right ordinate). The cyan arrow indicates the value of  $W_b^*$  ( $1.83 \text{ m s}^{-1}$ ), which represents the 86th percentile of the  $W$  spectra; b) Similar for the cloud droplet concentrations measured with the CCP-CDP probe. The cyan line indicates the  $N_d^*$  value ( $1207 \text{ cm}^{-3}$ ) at the 86th percentile in the  $N_d$  spectra. The indicated time is in UTC and shows the time of the first cloud penetration at cloud base and the total number of 1-s measured cloud data points.



1190 Figure 8.  $CCN_1$  (red dots) and  $CCN_2$  (black dots) measurements for a segment of flight AC17 on 27 September 2014. The  
 1195 abscissa shows the measurement time in UTC. The blue line indicates the altitude in meters above sea level and is labeled  
 on the left ordinate (as well as  $CCN_1$  and  $CCN_2$ ).  $S_1$  and  $S_2$  measurements in % are indicated by the orange and green  
 lines, respectively (both are labeled on the right ordinate). Cyan dots on the blue line indicate cloud penetrations (i.e.,  
 when cloud droplet concentrations are greater than  $20 \text{ cm}^{-3}$ ). In this case, cloud base heights were observed around 2,300  
 meters above ground.

1200

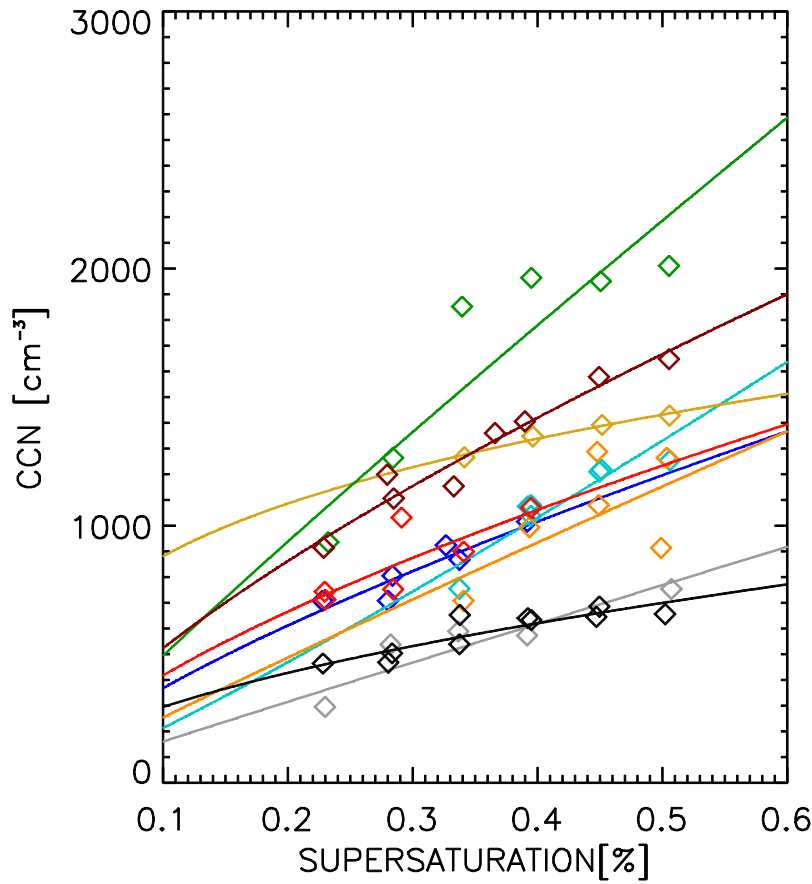
1205



1210 Figure 9. A comparison of the *CCN* spectra derived from the two *CCN* counter columns on board the HALO aircraft during flight AC17. Black (blue) smaller dots indicate *CCN*<sub>1</sub> (*CCN*<sub>2</sub>) measurements for each second. Large diamonds in black (blue) indicate the *mCCN*<sub>1</sub> (*mCCN*<sub>2</sub>) for each time step of measurements. The orange large diamonds indicate the *NCCN*<sub>2</sub> values, which are used to fit the power law equation of the group of measurements, which is shown at the lower right corner of the plot. The standard error for *CCN* spectra derived is shown at Table 2.

1215

1220



AC11 START: 14:58:21 (593 s)  
 CCN=1985.7 · S<sup>0.731</sup> R:0.91

AC11 START: 17:38:20 (710 s)  
 CCN=2927.4 · S<sup>1.137</sup> R:0.92

AC12 START: 15:56:00 (440 s)  
 CCN=1764.5 · S<sup>0.301</sup> R:0.99

AC13 START: 16:29:01 (722 s)  
 CCN=4145.4 · S<sup>0.922</sup> R:0.92

AC14 START: 15:21:40 (800 s)  
 CCN=1509.7 · S<sup>0.973</sup> R:0.89

AC15 START: 13:33:35 (555 s)  
 CCN=2209.0 · S<sup>0.930</sup> R:0.70

AC16 START: 20:21:40 (550 s)  
 CCN=1966.2 · S<sup>0.872</sup> R:0.82

AC17 START: 16:50:50 (831 s)  
 CCN=2743.2 · S<sup>0.719</sup> R:0.97

AC17 START: 19:38:20 (840 s)  
 CCN=1015.2 · S<sup>0.537</sup> R:0.90

1225

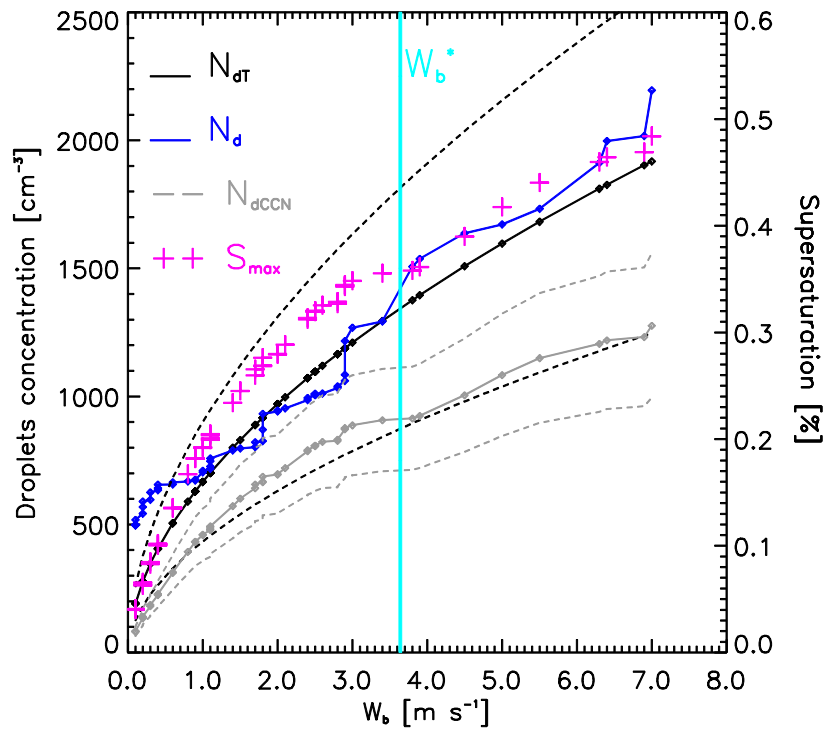
Figure 10. *CCN* spectra as measured on board the HALO aircraft during cloud profiling flights. Diamonds indicate the  $NCCN_2$  values, which are used to fit the power law equation of the group of measurements. The colors indicate the group of measurements and match the legend on the right side of the plot. The legend indicates the flight number; the initial time of group measurements; the period of measurements in seconds; the power law fit and the correlation coefficient of the data. The standard errors for each *CCN* spectra derived are shown at Table 2.

1230

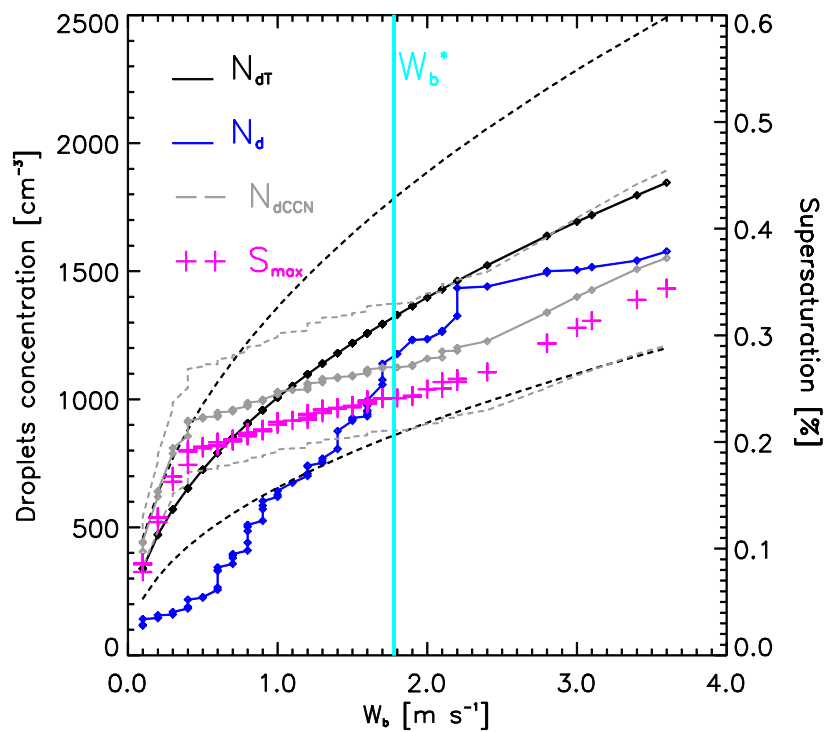
1235



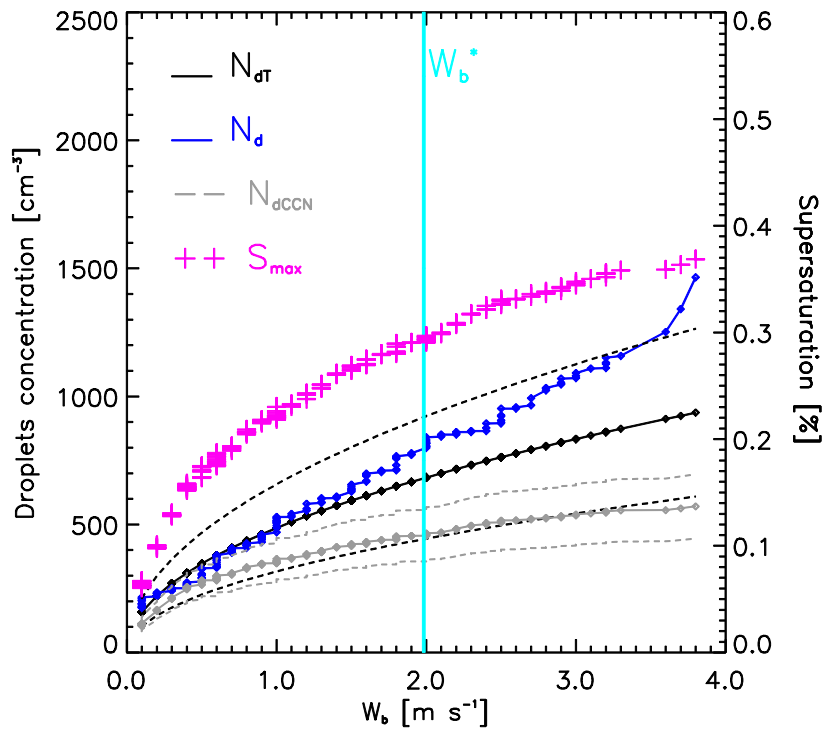
a) FLIGHT AC11 TIME: 17:52(56 s) CCN=2927.4 · S<sup>1.137</sup>



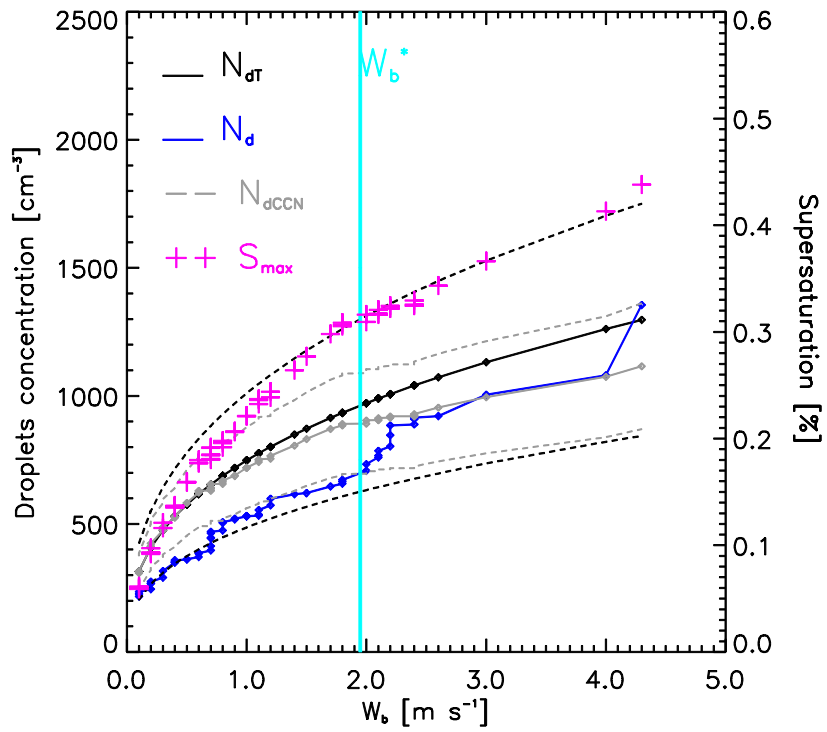
b) FLIGHT AC13 TIME: 16:50(72 s) CCN=4145.4 · S<sup>0.922</sup>



c) FLIGHT AC14 TIME: 15:06(184 s) CCN=1509.7 · S<sup>0.973</sup>



d) FLIGHT AC16 TIME: 20:10(58 s) CCN=1966.2 · S<sup>0.672</sup>



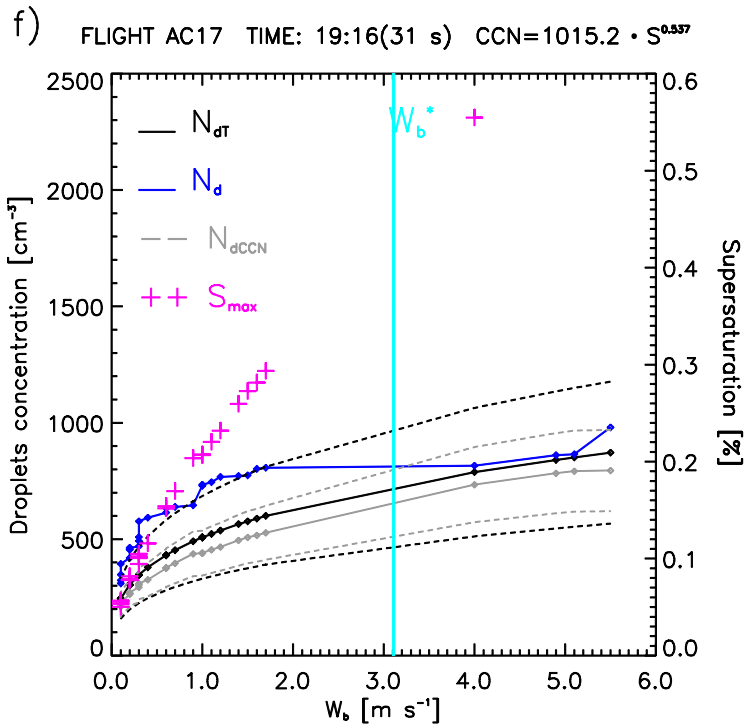
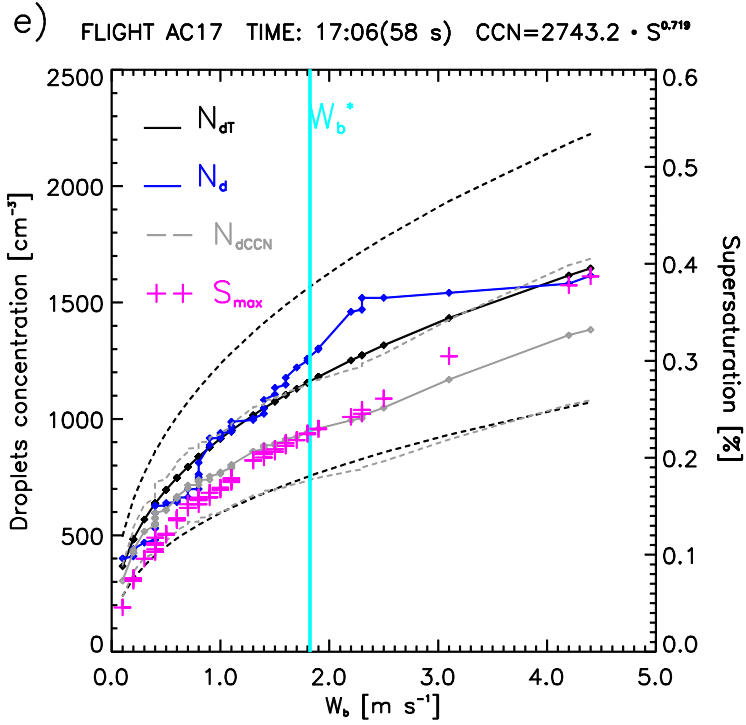
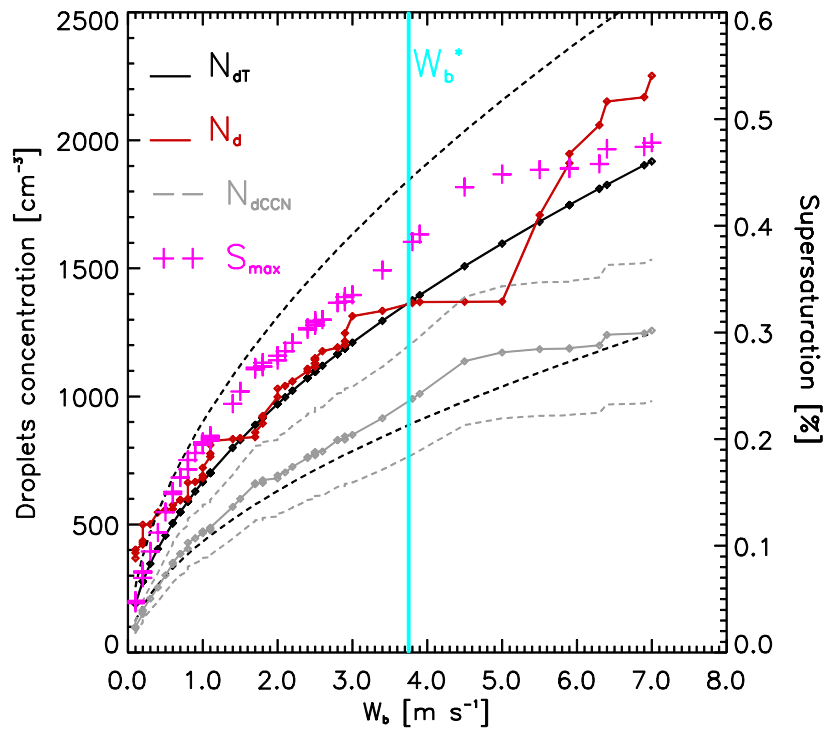


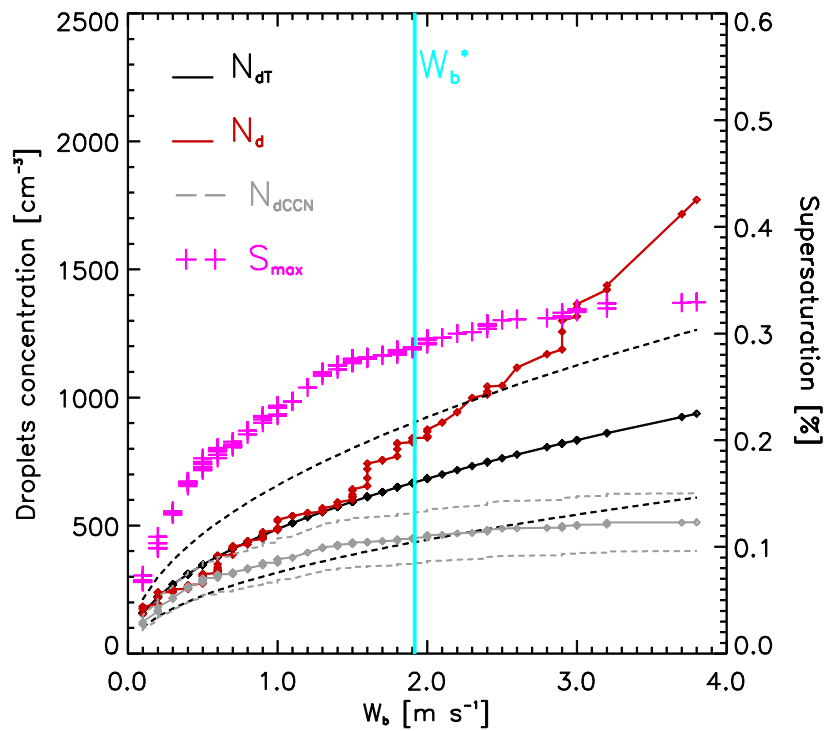
Figure 11a-f.  $N_{dCCN}$ ,  $S$ ,  $N_{dT}$  and  $N_d$  values are presented as a function of the cloud base updrafts ( $W_b$ ). This plot is based on the ‘probability matching method’ (PMM), using same percentiles for  $W_b$  and  $N_d$  ( $N_{dCCN}$  or  $N_{dT}$ ). The values of  $N_{dCCN}$ ,  $N_{dT}$  and  $N_d$  are shown the left y-axis, those of  $S$  on the right y-axis. The black dashed lines are the  $N_{dT}$  uncertainties. The gray solid (dashed) lines are the  $N_{dCCN}$  values (uncertainties). The effective updraft  $W_b^*$  for each flight segment is shown by the cyan line. The data are based on the CAS-DPOL probe. The time, period of measurements (sample size in seconds), and  $N_{CCN}(S)$  equation are shown on the top of the figures.

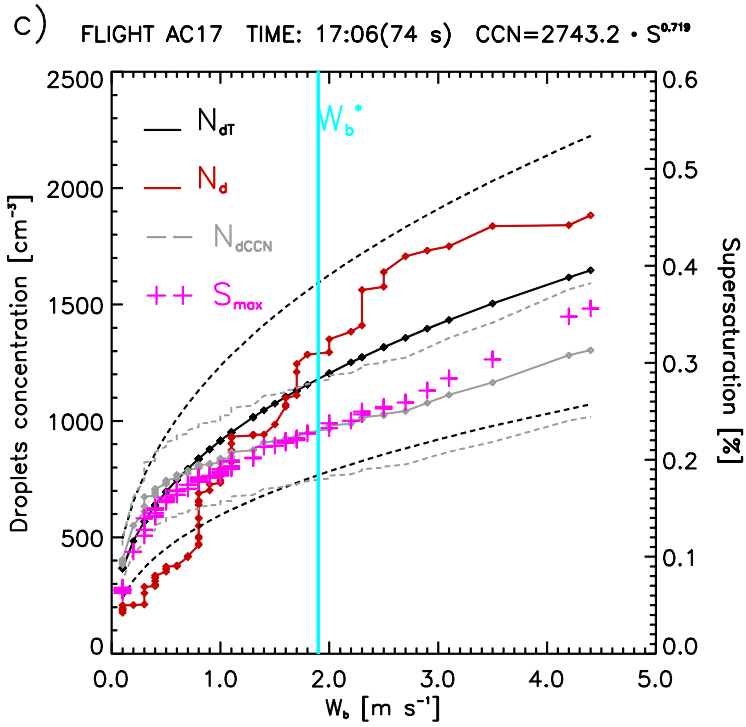
a) FLIGHT AC11 TIME: 17:52(61 s) CCN=2927.4 · S<sup>1.137</sup>



1255

b) FLIGHT AC14 TIME: 15:06(111 s) CCN=1509.7 · S<sup>0.973</sup>





1260

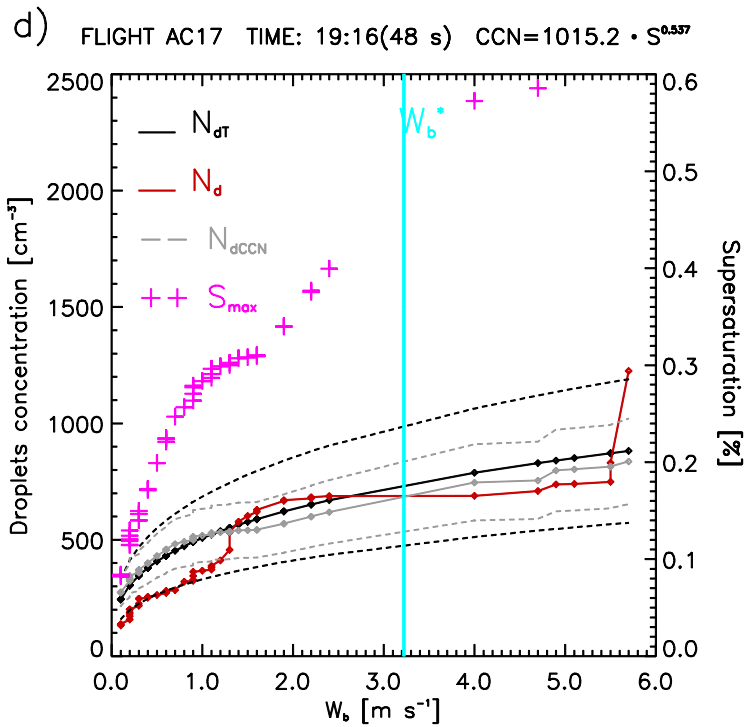


Figure 12a-d. Same as Figure 11 for the CCP-CDP probe. No data were available for flight AC16. The CCP-CDP malfunctioned in flight AC13 during the cloud base measurements.

1265

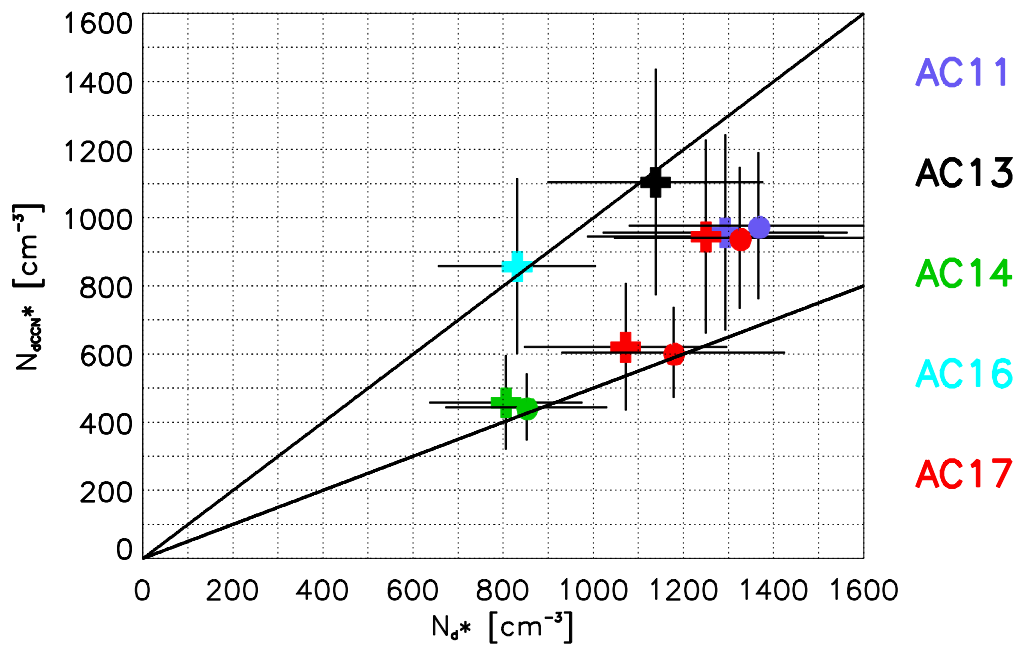
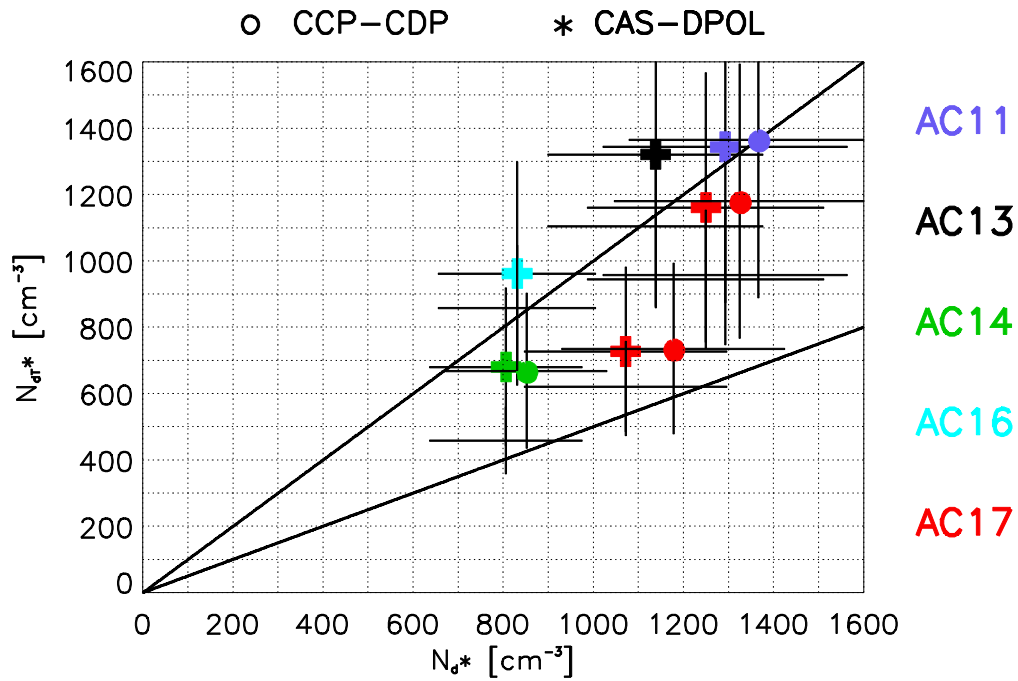
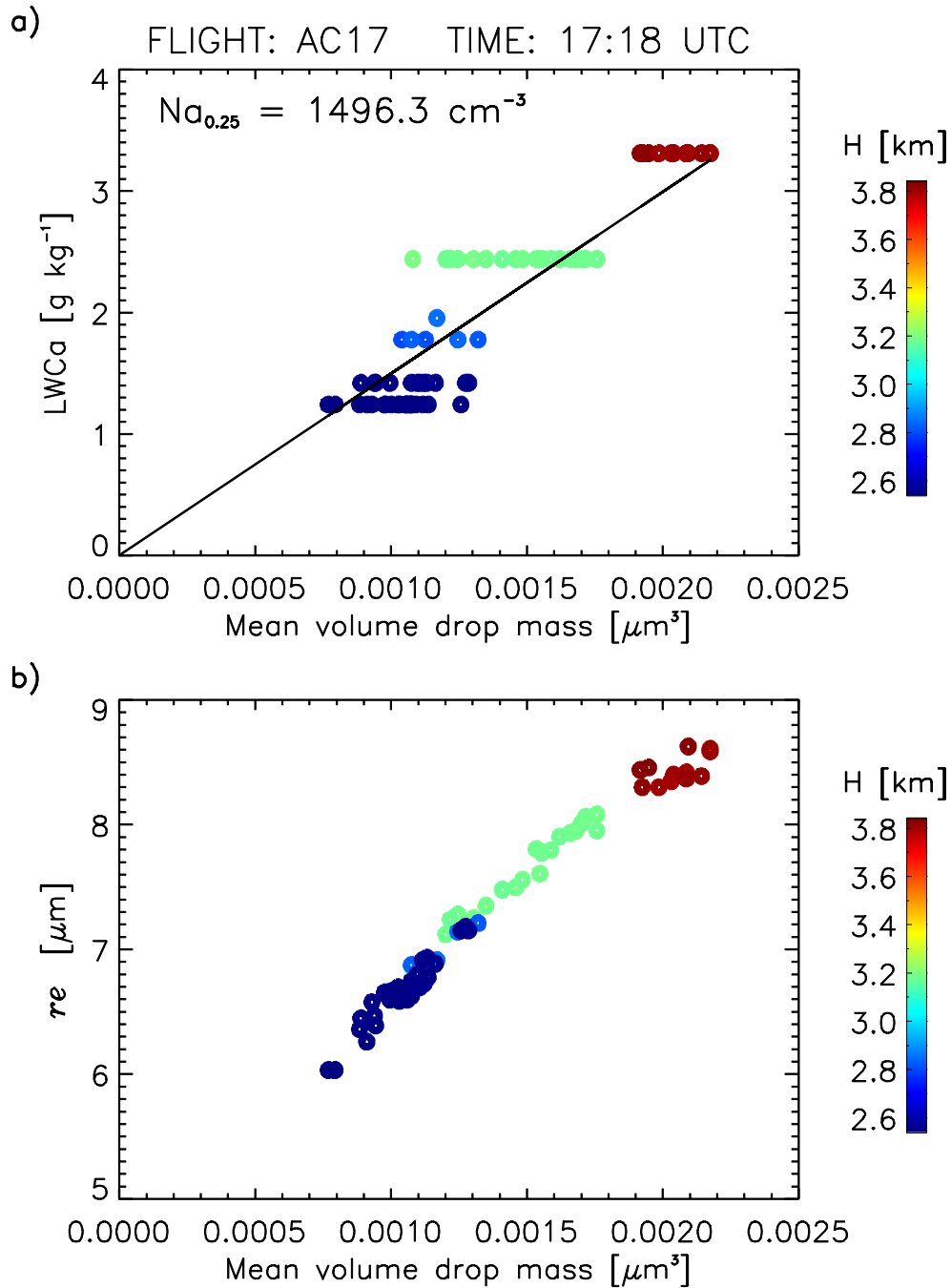
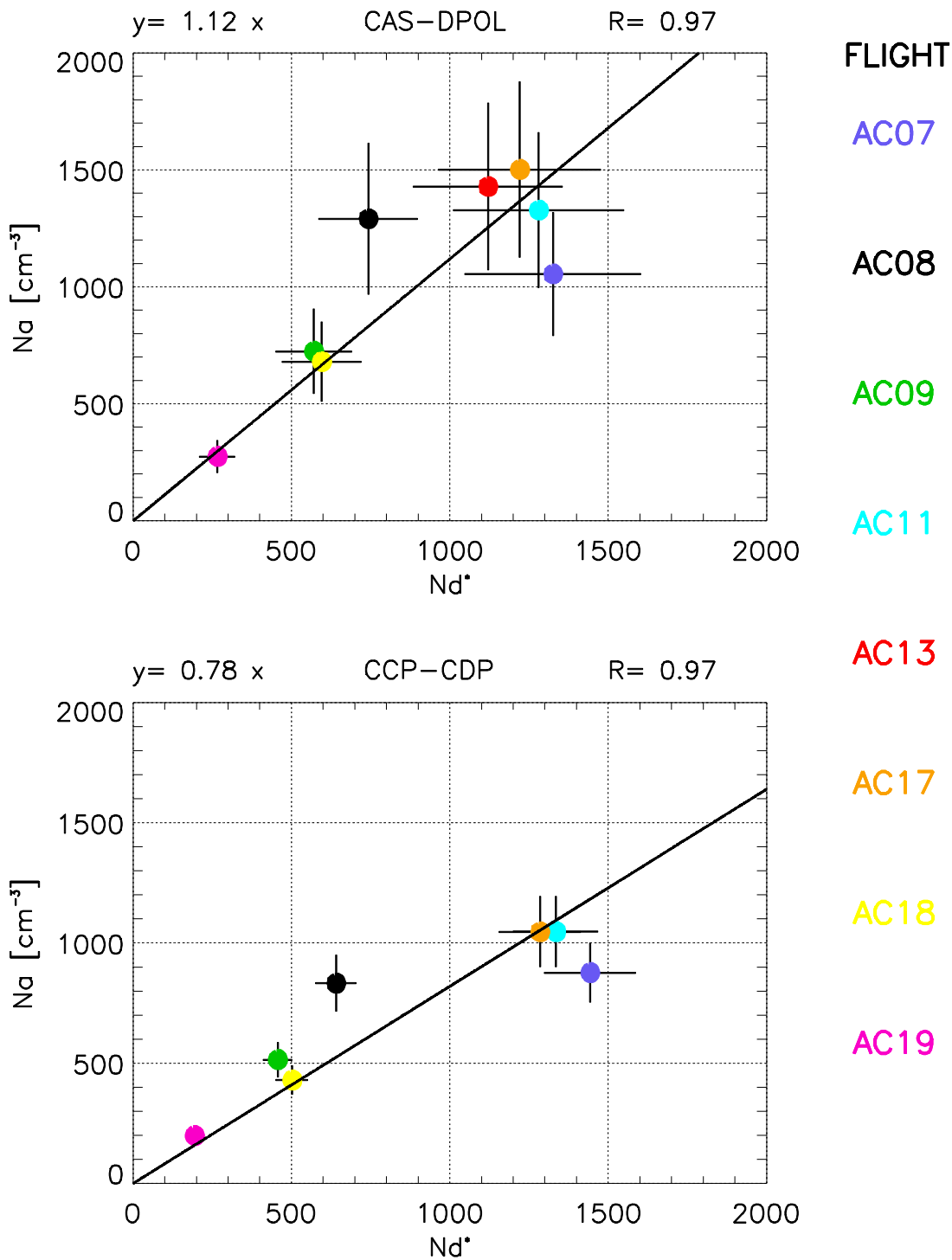


Figure 13. a)  $N_d^*$  versus  $N_{dT}^*$  calculated with  $W_b^*$  from cloud base data shown in Figures 11-12. The CAS-DPOL values are indicated by plus symbols (+) and the CCP-CDP values are indicated by circles (o). The colors indicate each flight segment (legend in the right side of the plot). Error bars indicate the uncertainties of variables estimates. Lines show the 1:1 and 1:2 relationships between  $N_{dT}^*$  versus  $N_d^*$  for each probe; b) Same for  $N_d^*$  versus  $N_{dCCN}^*$ .

1270



1275 **Figure 14 a).** Mean volume drop mass ( $M_v$ ) versus liquid water content from the CCP-CDP measurements for adiabatic fraction greater than 0.25 ( $LWC_a$ ). Values are shown with different colors labeled as a function of height in kilometers above sea level (indicated by the colorbar on the right side of the graphic). The slope of the linear equation is the estimated  $N_a$  (i.e.,  $1496 \text{ cm}^{-3}$ ); **b)**  $M_v$  versus  $r_e$  as a function of height in kilometers above sea level (indicated by the colorbar on the right side of the graphic).



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Figure 15.  $N_d^*$  versus  $N_a$  measured with CAS-DPOL and CCP-CDP (indicated on the top of panels) for profile flights during the ACRIDICON-CHUVA campaign. The color of the dots is associated with the flight number shown at the right side of the panels. Error bars indicates the uncertainties of variables estimates. The linear regression equation and the correlation coefficient  $R$  are shown in the top of each panel.

1285