



Cloud droplet number closure for tropical convective clouds during the ACRIDICON–CHUVA campaign

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Abstract.

The main objective of the ACRIDICON-CHUVA (Aerosol, Cloud, Precipitation, and Radiation Interactions and Dynamics of Convective Cloud Systems–Cloud Processes of the Main Precipitation Systems in Brazil: A Contribution to Cloud Resolving Modeling and to the Global Precipitation measurements) campaign in September 2014 was the investigation of aerosol-cloud-
5 interactions in the Amazon Basin. Cloud properties near cloud base of growing convective cumuli were characterized by cloud droplet size distribution measurements using a cloud combination probe (CCP) and a cloud and aerosol spectrometer (CAS-DPOL). In the current study, an adiabatic parcel model was used to perform cloud droplet number (N_d) closure studies for several flights in differently polluted air masses. Model input parameters included aerosol size distributions, measured with an ultra-high sensitive aerosol spectrometer (UHSAS), in combination with a condensation particle counter (CPC). Updraft
10 speeds (w) were measured near cloud base using a boom-mounted Rosemount model 858 AJ probe. To compare to model predictions, measured N_d and w were statistically matched based on equal percentiles of occurrence. Reasonable agreement between measured and predicted N_d was achieved when a particle hygroscopicity of $\kappa \sim 0.1$ is assumed. Similar closure results were obtained when the variability in the particle number concentration was taken into account. We conclude that N_d can be predicted using a single κ , and measured aerosol particle number concentration below cloud base when w is constrained
15 based on measurements. In accordance with previous adiabatic air parcel model studies, the largest disagreements between predicted and measured N_d were found when updraft speeds were high ($w > 2.5 \text{ m s}^{-1}$) or in the presence of a bimodal aerosol size distribution. We show that simplifying assumptions on κ might not be appropriate when the aerosol size distribution is



comprised of both distinct Aitken and accumulation modes, as predicted N_d clearly deviate from measured ones at $w \gtrsim 1 \text{ m s}^{-1}$ which points to a contribution of Aitken mode particles to N_d .

20 1 Introduction

Aerosol-cloud-interactions represent one of the largest uncertainties in our current understanding of the Earth's climate system, according to the latest Intergovernmental Panel on Climate Change (IPCC, 2013). Aerosols have a strong effect on cloud properties since cloud droplets form on cloud condensation nuclei (CCN) by condensation of water vapor. The uptake of water vapor by aerosol particles and the subsequent activation and growth of cloud droplets is described by the Köhler theory (Köhler, 25 1936). The Raoult effect (solute term) is commonly parameterized using the hygroscopicity parameter κ with values ranging from ~ 0.1 to ~ 0.9 for single components of atmospheric aerosol particles (Petters and Kreidenweis, 2007). The κ values of ambient aerosol particles are in the range of $\sim 0.09 < \kappa < 0.18$ for the complex mixtures of organic aerosols and $0.1 < \kappa < 0.3$ for biomass burning aerosols (e.g., Andreae and Rosenfeld, 2008; Carrico et al., 2010; Engelhart et al., 2012).

The Amazon Basin is a unique region to test our understanding of aerosol-cloud interactions in shallow and deep convective 30 clouds due to the large variability of aerosol composition and concentration during the dry and wet seasons (e.g., Andreae et al., 2004; Artaxo, 2002; Pöhlker et al., 2016, 2018). It is well established that the properties and dynamics of clouds can be fundamentally changed due to anthropogenic emissions into this pristine rainforest region (Pöhlker et al., 2018; Reutter et al., 2009; Roberts et al., 2003; Rosenfeld et al., 2008). To explore aerosol-cloud interactions, cloud microstructure and precipitation-forming processes in the Amazon rain forest, the ACRIDICON-CHUVA (Aerosol, Cloud, Precipitation, and 35 Radiation Interactions and Dynamics of Convective Cloud Systems - Cloud processes of tHe main precipitation systems in Brazil: A contribUtion to cloud resolVing modeling and to the GlobAl Precipitation Measurements) campaign with the HALO (High Altitude Long Range Research) aircraft was performed during the dry season in September 2014 (Wendisch et al., 2016).

Many CCN closure studies in various parts of the world have been performed to improve our understanding of the relationship between aerosol properties and their ability to form cloud droplets (e.g., Broekhuizen et al., 2006; Rissler et al., 2004; 40 Wang et al., 2008; Ervens et al., 2010). Such closure studies compare the predicted CCN number concentration (N_{CCN}) based on particle size and composition (hygroscopicity) to results from CCN measurements according to Köhler theory, i.e. for equilibrium conditions at different supersaturations in CCN counters. Much fewer studies compare predicted ($N_{d,p}$) and measured cloud droplet number concentrations ('cloud droplet number closure') ($N_{d,m}$), since often direct measurements or estimates of the updraft speeds (w) near cloud base are not available. Therefore, updraft speed is often used as a fitting parameter to match 45 $N_{d,m}$ (e.g., Anttila et al., 2012). Other studies used w distributions to yield a range of $N_{d,p}$ (Peng et al., 2005; Meskhidze et al., 2005; Hsieh et al., 2009; Chuang et al., 2000). Previous cloud droplet closure studies suggested that w is one of the most uncertain parameters leading to large uncertainties in the predictions of cloud droplet number concentrations (e.g., Conant et al., 2004; Fountoukis et al., 2007).

Braga et al. (2017) compared N_d at cloud bases measured during the ACRIDICON-CHUVA campaign to CCN number 50 concentration based on traditional parameterizations (Twomey, 1959). The current study represents an extension of this work,



in which we explicitly simulate condensational growth of aerosol particles from below cloud base up to a height of several meters above the level at which they grow to cloud droplet sizes. We compare measured cloud droplet number concentration ($N_{d,m}$) at cloud base of convective clouds in different air masses with those predicted ($N_{d,p}$) by an adiabatic parcel model (Ervens et al., 2005; Feingold and Heymsfield, 1992), using in situ measurements of aerosol size distributions as model input.

55 Model studies are performed to explore the sensitivities of particle hygroscopicity (κ), aerosol particle number concentration (N_a), and w to N_d .

2 Measurements of aerosol and cloud properties near cloud base

Aerosol and cloud measurements were performed during flights in convective cumulus clouds with lowest particle concentration during flight AC19, moderate concentrations during flights AC09 and AC18 and highest during flight AC07 in the southern and northern region of the Amazon Basin (Fig. S1; Tables S1-S4). Figure 1 shows the measurement region for the flights analyzed in this study. Cloud droplet number concentrations and size distributions were measured by a Cloud Combination Probe – Cloud Droplet Probe (CCP-CDP) and by a Cloud and Aerosol Spectrometer (CAS-DPOL) mounted onboard HALO (Wendisch et al., 2016). Cloud droplet number size distributions (DSDs) between $3\ \mu\text{m}$ and $50\ \mu\text{m}$ in diameter were measured at a temporal resolution of 1 s by the CAS-DPOL and CCP-CDP probes (Baumgardner et al., 2011; Voigt et al., 2010, 2011; 65 Kleine et al., 2018; Wendisch and Brenguier, 2013). Each DSD spectrum represents 1 s of flight path (covering between 70 m to 120 m of horizontal distance for the aircraft speed at cloud bases). From the DSDs, the droplet number concentrations were derived by size integration. Braga et al. (2017) showed that both probes were in agreement within the measurement uncertainties ($\pm \sim 30\%$) with respect to $N_{d,m}$ at cloud bases. A positive bias of $\sim 20\%$ was found for averaged CAS-DPOL measurements of $N_{d,m}$ at cloud bases in comparison with those measured with CCP-CDP. Cloud passes were defined for conditions, under which 70 the number droplet concentration (i.e. particles with diameter larger than $3\ \mu\text{m}$) exceeded $20\ \text{cm}^{-3}$. This categorization was applied to avoid cloud passes well mixed with environment air and counts of haze particles. The HALO aircraft was equipped with a meteorological sensor system (BAasic HALO Measurement And Sensor System; BAHAMAS) located at the nose of the aircraft. Updraft speeds at cloud base were measured in a range of $0.1\ \text{m s}^{-1} \leq w \leq 6\ \text{m s}^{-1}$, using a boom-mounted Rosemount model 858 AJ air velocity probe. The uncertainties in measured w are $\Delta w < 0.2\ \text{m s}^{-1}$ for $w < 5\ \text{m s}^{-1}$ and $\Delta w \approx 0.25\ \text{m s}^{-1}$ for 75 $w > 5\ \text{m s}^{-1}$. Further details on the uncertainties of w measurements are described by Mallaun et al. (2015).

The particle number concentration with diameter $> 20\ \text{nm}$ (N_a) below cloud base were measured using the Aerosol Measurement System (AMETYST); the uncertainty of these measurements is estimated to be 10 % (Andreae et al., 2018). Particle number concentrations ranged from $\sim 500\ \text{cm}^{-3}$ to $\sim 2000\ \text{cm}^{-3}$. Dry particle size distributions were measured by an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS) covering a size range between 60 nm and 600 nm (Brock et al., 2011; Cai 80 et al., 2008) with an estimated uncertainty of $\sim 20\%$ (Andreae et al., 2018). The particle size distributions for all flights were fitted by lognormal distributions with typical parameters for an accumulation mode with a mean diameter between 136 nm and 147 nm (Figure S1; Tables S1-S4). For flight AC19, an additional Aitken mode was inferred with a mean diameter of 37 nm



to match the observed width and N_a of the aerosol size distribution. The uncertainty of these size distribution measurements is estimated to be $\pm 30\%$. These aerosol size distributions were used as input to the adiabatic parcel model (Section 3.2).

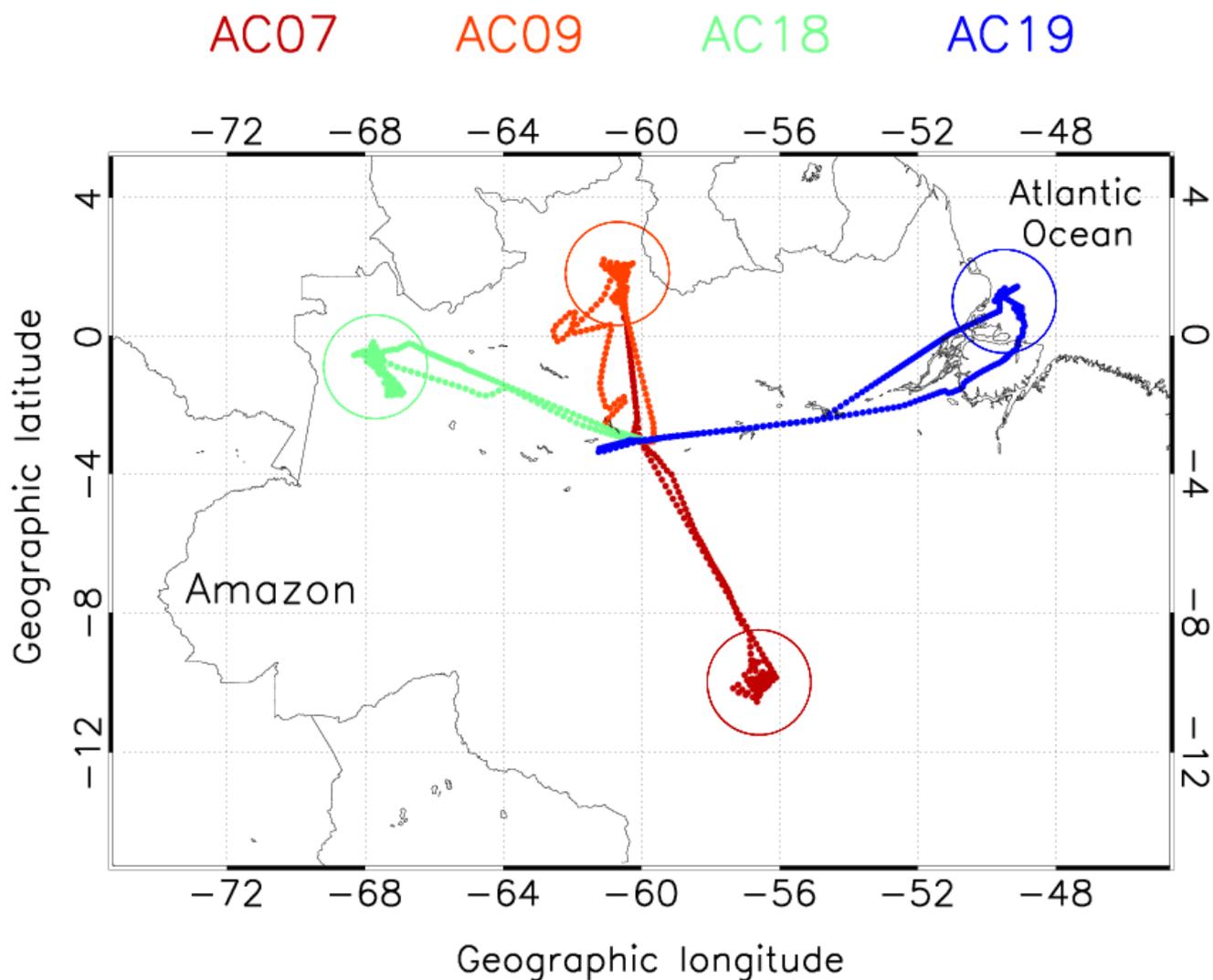


Figure 1. HALO flight tracks of the four different flights (AC07, AC09, AC18, AC19) used in this study during the ACRIDICON-CHUVA campaign. Colored circles indicate the region of cloud base measurements during each flight.



85 3 Methodology

3.1 Probability matching method (PMM): Pairing measured updraft speeds (w) and droplet number concentrations ($N_{d,m}$)

As w is highly variable in clouds and is measured independently from $N_{d,m}$, we apply the “Probability Matching Method” - PMM (Haddad and Rosenfeld, 1997) to statistically determine the most probable combinations of $N_{d,m}$ and w values using the same percentiles of their occurrence. The PMM analysis is based on the assumption that these two related variables increase monotonically with each other. Measured $N_{d,m}$ and w values were sorted in ascending order and the most likely w value was assigned to $N_{d,m}$, measured during the four flights. To avoid biases caused by outlier measurements, $N_{d,m}$ above the 97.5th and below the 2.5th percentile were removed. Only cloud passes with positive w were considered in the analysis. Braga et al. (2017) have shown that this method can be used to find the best agreement between measured and estimated N_d at cloud base as a function of w . In the current study, PMM analysis is used to compare the $N_{d,m}$ near cloud base and its assigned w with $N_{d,p}$ at a constant w in the model.

3.2 Adiabatic cloud parcel model

3.2.1 Model description and simulations

The adiabatic parcel model describes the growth of aerosol particles by water vapor uptake on a moving mass grid (Ervens et al., 2005; Feingold and Heymsfield, 1992). The air parcel is described to rise with a constant w below and inside of the cloud. Saturation with respect to water vapor in the air parcel is calculated based on the standard thermodynamic equations for adiabatic conditions as a function of w and particle properties (N_a , particle sizes and hygroscopicity) (Pruppacher and Klett, 1997). It is assumed that the aerosol particles are internally mixed with identical hygroscopicity (κ) of all particles. Simulations are performed up to a height of 70 m above the level of predicted maximum supersaturation. The initial conditions for the model simulations are summarized in Tables S1-S4. Particles that exceed a diameter of 3 μm are defined as droplets; this definition allows a direct comparison of $N_{d,p}$ and $N_{d,m}$. Collision/coalescence processes are not considered. Sensitivity studies are performed for the observed ranges of w and $N_a \pm 30\%$ and assumed range of $0.02 \leq \kappa \leq 1$ to identify parameter ranges for which droplet closure can be achieved.

3.2.2 Determination of in-cloud height to compare $N_{d,m}$ and $N_{d,p}$

The measurements were performed at approximately constant altitude during each research flight. However, this height might represent different levels in relation to cloud base and to the level of maximum supersaturation, depending on updraft speed and turbulence in cloud. In order to determine the height at which $N_{d,m}$ and $N_{d,p}$ should be compared, simulations were performed using the measured aerosol particle size distributions and an assumed hygroscopicity of $\kappa = 0.1$, together with w measured at cloud base. Under adiabatic conditions, $N_{d,p}$ is approximately constant at ~ 20 m above the level of the maximum supersaturation S_{max} (Fig.S2).

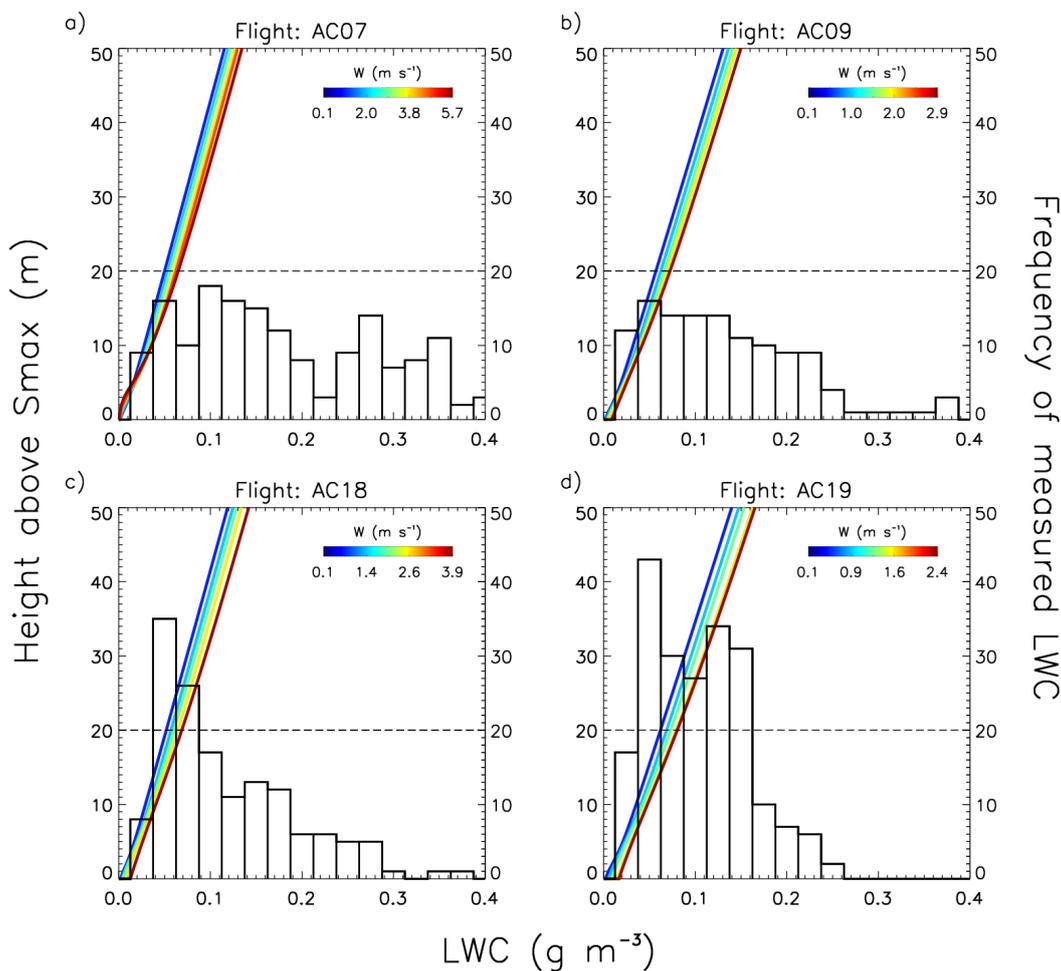


Figure 2. Predicted LWC [$g\ m^{-3}$] as a function of heights above the level of S_{max} [left axis] and w (lines, color-coded by w [$m\ s^{-1}$]) for flights a) AC07, b) AC09, c) AC18, d) AC19. The vertical bars indicate the number of cloud passes (with a temporal resolution of 1 s) as a function of the measured LWC by CAS-DPOL and CCP-CDP in cloud [right axis]. The dashed line denotes the level of 20 m above predicted maximum supersaturation, for which $N_{d,p}$ is predicted.



Figure 2 shows the values of predicted liquid water content (LWC) from the same simulations as a function of height above S_{max} for the four flights. Overlaid on the model results are the frequencies of measured LWC by the cloud probes near cloud base. The measured LWC represents the cumulative mass size distribution. For AC09, AC18 and AC19, the model predictions match the most frequently measured LWC at ~ 20 m above the S_{max} level. This height level might represent slightly different absolute heights above the surface and the level of saturation (RH = 100%) (Fig. S3). For AC07, the LWC frequency distribution is very flat and leads to ambiguity of the height in cloud where predicted and measured LWC coincide. However, since we focus our discussion in the following on the comparison of $N_{d,m}$ and $N_{d,p}$, we perform our analysis for a height of 20 m above S_{max} for all flights, as above this height cloud droplet number is not predicted to change.

4 Results and Discussion

4.1 Constraining aerosol hygroscopicity (κ) based on N_d and w

Figure 3 shows the range of $N_{d,m}$ as a function of w as determined by the PMM method (Section 3.1); the symbols indicate $N_{d,m}$ from CAS-DPOL (red symbols) and CCP-CDP (blue symbols). The lines indicate model predictions for the assumption of $\kappa = 0.05$, $\kappa = 0.1$ and $\kappa = 0.3$ (dashed, dotted, solid lines, respectively) for up to 38 w values for each flight, covering the measured w range. For all flights, $N_{d,m}$ are reasonably reproduced by the model assuming a particle hygroscopicity of $\kappa \sim 0.1$; $N_{d,p}$ are closer to the measured values from CCP-CDP assuming a slightly lower κ , whereas $N_{d,m}$ from CAS-DPOL indicate a slightly higher κ . However, these deviations are within the uncertainty range of the cloud probe measurements, i.e., $\sim 10\%$ and $\sim 20\%$ for CCP-CDP and CAS-DPOL, respectively (Braga et al., 2017).

Generally, best agreement between measured and predicted cloud droplet number concentration is obtained for low w during all flights. However, the value of w , above which the model predictions deviate from measurements varies among the flights: For continental clouds as encountered during AC07, AC18 and AC09, the model results agree well with observations for $w \lesssim 2.5$ m s^{-1} . At higher w , $N_{d,m}$ shows a much stronger increase with w than predicted by the model. For AC19, i.e. above the ocean, this trend is even obvious for $w \gtrsim 0.5$ m s^{-1} .

The results in Figure 3 imply that the assumption of an internally mixed aerosol population with moderate hygroscopicity ($\kappa \sim 0.1$) is justified to reproduce $N_{d,m}$ for flights AC07, AC09 and AC18 for wide ranges of updraft speeds (0.1 $\text{m s}^{-1} \leq w \leq 2.5$ m s^{-1}). A similar κ value has been suggested previously for similar air masses during the dry season in the Amazon Basin (e.g., Pöhlker et al., 2016, 2018). In these prior studies κ was constrained to a value of ~ 0.1 based on size-resolved CCN measurements and measurements of the aerosol chemical composition, dominated by an aged organic fraction. Our results confirm this κ value that is representative of internally mixed aerosol particle populations during the dry season in the Amazon Basin, which is influenced by fresh and aged biomass burning aerosol from Amazon and Africa (Holanda et al., 2020).

The systematic bias in $N_{d,p}$ suggests that the adiabatic model might not be suitable for high updraft conditions. Under such conditions, entrainment of additional aerosol particles from air masses surrounding the cloud could explain the observed trend in $N_{d,m}$ which would not be captured by the model. Depending on the conditions, entrainment has been shown to lead also to the opposite trend, i.e., to the decrease of N_d (Calmer et al., 2019).

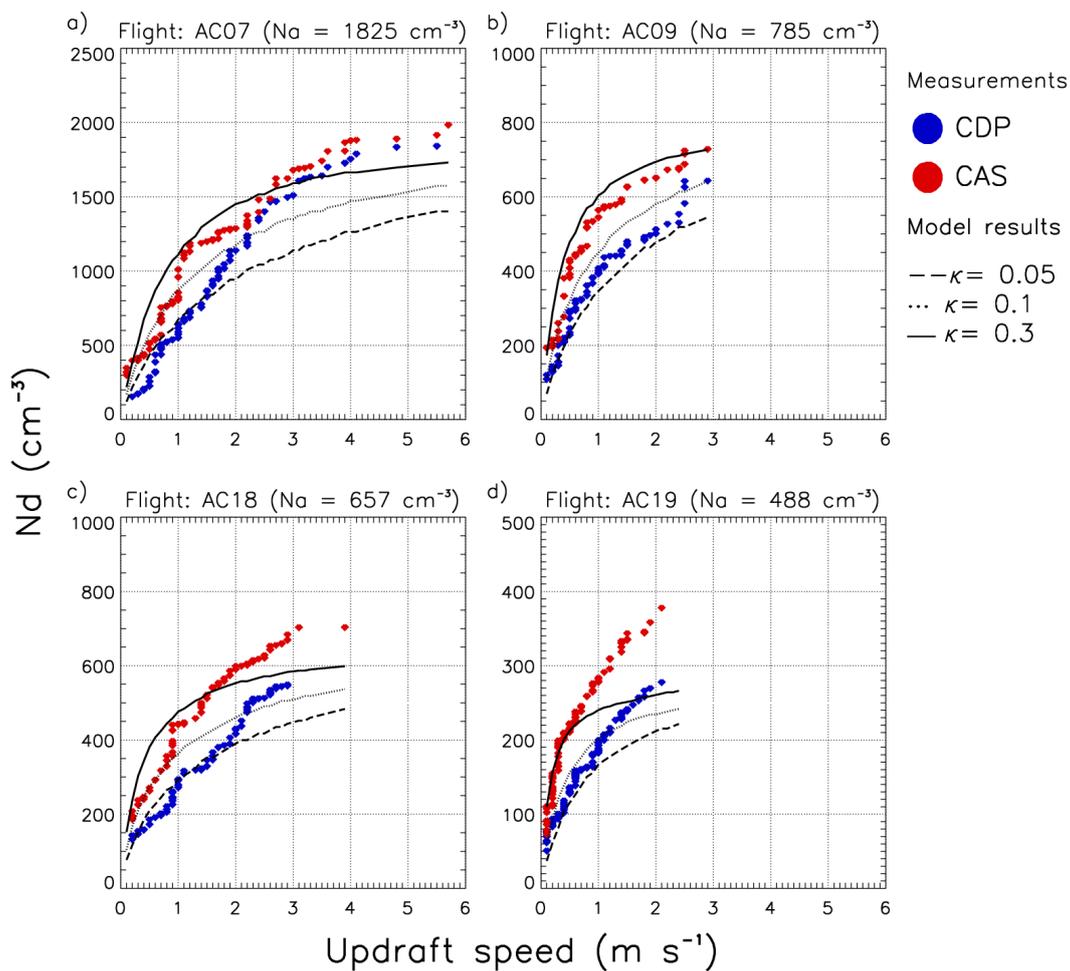


Figure 3. Cloud droplet number concentration (N_d) as a function of updraft speed near cloud base of convective clouds during flights: a) AC07, b) AC09, c) AC18 and d) AC19. The measured updraft speeds are based on the “probability matching method” (PMM) using the same percentiles for updraft speed and $N_{d,m}$ (Section 3.1). The red and blue symbols represent $N_{d,m}$ near cloud base with the CAS-DPOL and CCP-CDP probes, respectively. Measurement uncertainties are $\sim 20\%$ and $\sim 10\%$ for CAS-DPOL and CCP-CDP data (Braga et al. (2017)). The dashed, dotted and solid lines show $N_{d,p}$ assuming $\kappa = 0.05$, $\kappa = 0.1$ and $\kappa = 0.3$, respectively.



While also different hygroscopicities of particles activated at different w might explain the trends in Figure 3a-c, it seems
150 unlikely that during the same flight, aerosol populations with very different hygroscopicity were collected. While the hygro-
scopicity of particles could possibly change near the cloud base, e.g. due to dissolution of soluble compounds, this effect would
result in the opposite trend as predicted, i.e., a higher $N_{d,p}$ at low w when particles dissolve over longer time scales. The result-
ing curves of $N_{d,p}$ as a function of w would be even flatter than shown in Figure 3, as opposed to the steep increase in $N_{d,m}$.
Thus, a significant role of such composition effects can likely be excluded.

155 The air masses below cloud encountered during flight AC19 were mostly impacted by marine air leading to a bi-modal
aerosol size distribution with low $N_{d,m}$ (Figure S1d). For this flight, the cloud droplet closure is worse as compared to the
reasonable agreement for the other three cases. Not only is the absolute difference between $N_{d,m}$ and $N_{d,p}$ relatively larger
(Figure 3d), but also the trend of $N_{d,m}$ with w cannot be well reproduced: while at $w < 0.5 \text{ m s}^{-1}$, the range of $N_{d,p}$ agrees well
with $N_{d,m}$ above this threshold, the model strongly underestimates the droplet number concentration even for $\kappa = 0.3$ (Figure
160 3d).

The measured aerosol size distribution during flights AC19 differed significantly from the other ones (Figure S1) because of
(i) low N_a , and (ii) a distinct Aitken mode (mean diameter 37 nm) that comprised $\sim 46\%$ of the particle number concentration.
At such low N_a , the maximum supersaturation in the clouds is relatively high so that at sufficiently high w Aitken mode
particles (diameter $\leq 80 \text{ nm}$) may be activated into cloud droplets and contribute to N_d . The chemical composition of Aitken
165 mode particles often differs significantly from that of accumulation mode particles, which are more aged and internally mixed
(e.g., Pöhlker et al. (2018); Wex et al. (2016)). Continental Aitken mode particles usually exhibit a lower hygroscopicity than
accumulation mode particles (McFiggans et al., 2006). Marine particles often show similar hygroscopicity in both Aitken and
accumulation modes (Kristensen et al., 2016; Wex et al., 2016). In our model, the same κ value for both modes is assumed. We
did not perform any further sensitivity studies to constrain different values for the hygroscopicity or mixing state in the two
170 modes. Depending on these mode-dependent aerosol characteristics, the contribution of Aitken mode particles to N_d at higher
updraft speeds might result in a flatter or steeper slope than predicted in Figure 3d.

In general, the observed trends of N_d with w confirm results from previous sensitivity studies that have shown that with
increasing w , changes in N_d become small and, thus, sensitivity of N_d to κ and w decreases (Ervens et al., 2005; Reutter
et al., 2009; Fountoukis et al., 2007). In these studies, it was demonstrated that at high w , the activated particle fraction is
175 sufficiently high that additional activation does not lead to a significant increase in activated particles, and, thus, the sensitivity
to N_d becomes small. In these studies either only an accumulation mode was considered, or N_d closure studies were performed
for situations with low w and/or fairly small Aitken mode particles ($< 40 \text{ nm}$). Based on a sensitivity study over wide ranges
of Aitken mode particle properties, Anttila and Kerminen (2007) concluded that the high sensitivities of N_d to the chemical
composition of Aitken mode particles might affect cloud properties. None of the previous studies showed the extent to which
180 presence of an Aitken mode might significantly affect N_d in convective clouds. Figure 3d clearly shows that the simplified
assumption of a single κ is not appropriate to infer $N_{d,p}$ for low aerosol loading and when N_a of the accumulation and Aitken
modes are comparable.

The sensitivities to N_a for relative contributions of Aitken and accumulation modes to total N_a will be systematically explored in a future study to identify conditions under which Aitken mode particles may impact cloud properties near cloud base.

185 4.2 Influence of aerosol number concentration (N_a) on predicted N_d

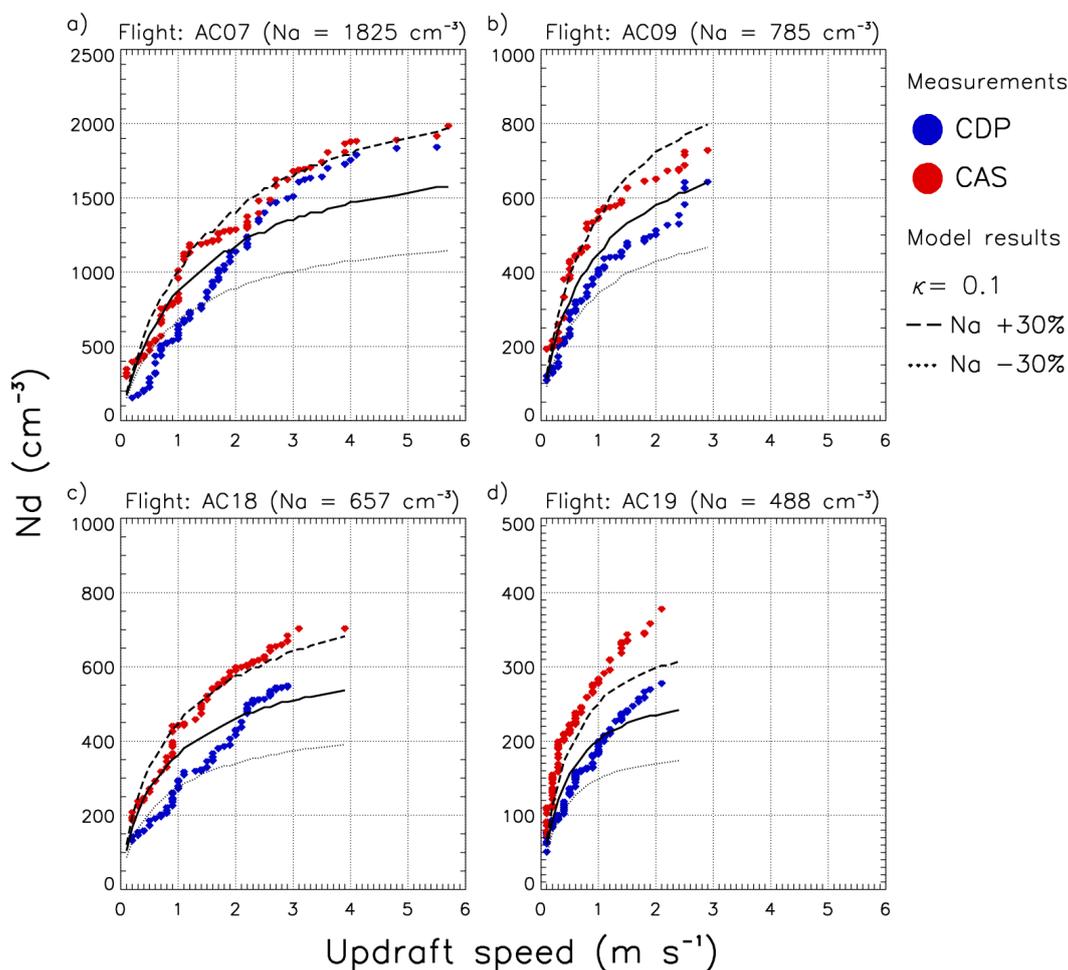


Figure 4. Cloud droplet number concentration (N_d) as a function of the updraft speed at cloud base of convective clouds during flights: a) AC07, b) AC09, c) AC18 and d) AC19. This plot is based on the “probability matching method” (PMM) using the same percentiles for updraft speed and $N_{d,m}$. The red (blue) dots are measured values at cloud base with the CAS-DPOL (CCP-CDP) probe. The measurement uncertainties are $\sim 20\%$ and $\sim 10\%$ for CAS-DPOL and CCP-CDP data. The solid black lines are simulated values with the parcel model assuming $\kappa = 0.1$. The dashed lines are the variability range of the simulated values ($N_{d,p}$) for the uncertainty range of N_a measurements ($\pm 30\%$).



The measurements of N_a were associated with uncertainties of $\pm \sim 30\%$. In order to account for this uncertainty, $N_{d,m}$ and $N_{d,p}$ are compared for all flights, using N_a (Figure S1), reduced by -30% and increased by $+30\%$ as model input, respectively. A hygroscopicity of $\kappa = 0.1$, as an average value based on the results in Section 4.1. was assumed. In Figure 4, the comparison of measured and predicted N_d is shown assuming these ranges of N_a . The solid lines repeat the same values as in Figure 3 ($\kappa =$
190 0.1); the dashed and dotted lines denote $N_{d,p}$ using the higher and lower input N_a . Similar to the findings in Figure 3, for flights AC07, AC09 and AC18, $N_{d,p}$ is within the range of $N_{d,m}$ for the assumed model parameter space. Also the curves for $N_{d,p}$ as a function of w exhibit the same shape as predicted for a variation in κ . The agreement between measurements and model results decreases with increasing w .

The closure results in Figure 4 show that $N_{d,m}$ can be reproduced reasonably by the model over wide w ranges if the
195 uncertainties in N_a measurements ($\pm 30\%$) are taken into account, and an average hygroscopicity of $\kappa = 0.1$ is assumed. For both sets of simulations, i.e. varying N_a or κ , the agreement between model and measurements is best at low w . At $w \gtrsim 2 \text{ m s}^{-1}$, the $N_{d,p}$ curves flatten suggesting a decreasing sensitivity of $N_{d,p}$ above this w threshold. Unlike the trends in $N_{d,p}$ for different κ values, the lines for different N_a keep diverging with increasing w . Thus, the sensitivity of $N_{d,p}$ to N_a is predicted to remain high, nearly independent of w . Similar to the conclusions on Figure 3d, the relatively larger discrepancies in both the
200 trends and absolute differences in $N_{d,p}$ and $N_{d,m}$ for AC19 (Figure 4d) compared to the closure results for the other three flights, points again to a potential role of Aitken mode particles in affecting N_d .

5 Summary and conclusions

Airborne measurements of cloud droplet number concentrations ($N_{d,m}$), aerosol particle size distributions and updraft speeds (w) near cloud base were performed during the ACRIDICON-CHUVA campaign in September 2014. Using an adiabatic air
205 parcel model, the effects of aerosol particle number concentration (N_a) and hygroscopicity (κ) on predicted cloud droplet number concentrations ($N_{d,p}$) near cloud bases of growing convective cumuli formed over the Amazon were explored. Data from aerosol and cloud probes onboard HALO were used as model input for this cloud droplet closure analysis. Model results for four different scenarios in terms of aerosol loading and size distributions and w confirm previously suggested values of the hygroscopicity parameter κ to reasonably predict N_d for most conditions: best N_d closure is achieved for $\kappa \sim 0.1$ comparing
210 to CCP-CDP and CAS-DPOL measurements, respectively. This droplet closure study represents a complementary approach to constrain CCN hygroscopicity, in addition to previous studies in this region, during which a similar κ range (0.1–0.35) was determined for aerosol in the Amazon Basin, and a range of $0.1 < \kappa < 0.9$ above the ocean, based on CCN measurements and detailed analysis of chemical composition (Pöhlker et al., 2016, 2018; Thalman et al., 2017; Wex et al., 2016). The comparison between predicted and measured N_d showed largest discrepancies at high updraft speeds ($w > 2.5 \text{ m s}^{-1}$), which
215 may be explained by non-adiabaticity and entrainment of aerosol particles in convective clouds. The variability of predicted N_d due to inferred κ ranges confirm trends from previous sensitivity studies for one-modal aerosol size distributions: The sensitivity to N_d decreases with increasing w , i.e. when the activated fraction is large and activation of additional smaller particles increases N_d only to a small extent (Ervens et al., 2005; Reutter et al., 2009; Cecchini et al., 2017; Pardo et al., 2019).



220 Uncertainties in N_a measurements ($\pm 30\%$) translate into similar differences in predicted droplet number concentration as
those for different κ values, in particular at low w . In previous cloud droplet number closure studies, composition effects, such
as slow dissolution of soluble compounds (Asa-Awuku and Nenes, 2007), reduced surface tension or variation of the water
mass accommodation coefficient (Conant et al., 2004) have been inferred to explain observed droplet number concentrations.
Our analysis shows that measurement uncertainties in basic aerosol properties might equally explain such differences. In the
225 case of a two-modal aerosol size distribution with distinct Aitken and accumulation modes (flight AC19), predicted N_d was
significantly smaller than the measured one for $w \gtrsim 0.5 \text{ m s}^{-1}$. A similar threshold, above which the Aitken mode might affect
 N_d was suggested in a recent study on marine stratocumulus clouds (Schulze et al., 2020). It may be concluded that the ratio
of the number concentrations of the Aitken and accumulation modes and their κ values might influence cloud properties near
cloud base differently than for one-modal aerosol size distributions. Sensitivity studies of cloud properties to Aitken mode
aerosol properties (N_a, κ) are warranted to identify conditions, under which they might affect aerosol-cloud interactions.

230 *Data availability.* The data used in this study can be found at <https://halo-db.pa.op.dlr.de/mission/5>.

Author contributions. RCB, BE, MLP led the analyses and the manuscript preparation. The measurements of aerosol and cloud properties
were conducted and analyzed by RCB, DF, BAH, TJ, OOK, CP, DS, CV, AW, MLP. The measurements were led by MOA, MW, UP. The
modeling studies were performed and interpreted by RCB, BE, DR, JDF, OL, LHP, LATM, MLP.

Competing interests. The authors declare that they have no conflict of interest.

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