Author response to referee comments

Referee 1

We thank the referee for the detailed comments and the good suggestions for improving the paper. We have addressed all comments as listed below which significantly improved our manuscript. Referee comments are in black, our responses in blue and manuscript text in *italic* and new text in *red*.

General changes in the manuscript:

1. We clarified how we constrain the measurements on board the HALO aircraft to investigate the relationship between w, N_a, N_d and κ using airborne data and model simulations. This is the first time that such measurements are performed with the proposed methodology ('Probability Matching Method' - PMM). The results from PMM analysis have shown agreement with previous studies and represents a complementary approach in which w, N_a, N_d can be used to constrain CCN hygroscopicity;
2. We revised the abstract and conclusion section to more clearly highlight the new findings and approaches in the current study.
3. More details on the measurements of aerosol and droplet concentrations below and at cloud bases of growing convective cumuli on board the HALO aircraft are are given in terms of time, location and uncertainties (Section 2 and supplemental information);
4. We added statistical parameters to quantify the agreement in the droplet closure.
5. We added a new *Section 4.3: Sensitivities of N_d predictions to w, N_a and κ where we discuss sensitivities of cloud droplet number concentration to κ, N_a and w ($\xi(\kappa)$, $\xi(N_a)$ and $\xi(w)$) in the context of previous sensitivity studies.*

Below you find our specific responses to the referee comments.

1 Referee Comment: A number of questions remain about the measurements used in the study. Why are the CDP and CAS droplet measurements so different? What measurements were actually used during the flight? Was the entire flight averaged? Were cloud edges excluded? Were the aerosol measurements collected relatively close to the cloud droplet measurements?

Author response: We substantially extended Section 2 (Aircraft measurements) and added specific sections on aerosol and cloud measurements

2 Aircraft Measurements

~~of aerosol and cloud properties near cloud base~~

The measurements were performed aboard the High Altitude LOng Range aircraft (HALO), a modified business jet G550 (manufactured by Gulfstream, Savannah, USA). In situ meteorological and avionics data, such as the vertical velocity, were obtained at 1 Hz from the BAsic HALO Measurement And Sensor System (BAHAMAS). A boom-mounted Rosemount model 858 AJ air velocity probe was used to measure the updraft velocity with BAHAMAS, measuring in a range of 0.1 m s^{-1} $\leq$ w $\leq$ 6 m s^{-1}. The uncertainties in measured w are $\Delta w < 0.2$ m s^{-1} for $w < 5$ m s^{-1} and $\Delta w \approx 0.25$ m s^{-1} for $w > 5$ m s^{-1}. Further details on the uncertainties of w measurements are described by Mallaun et al. (2015). The measurements took place over the Amazon

Basin and over the western tropical Atlantic in September 2014 during the ACRIDICON–CHUVA campaign (Wendisch et al., 2016).

Figure 1a shows the measurement region for the flights analyzed in this study (the flights are labelled with 'AC' and a running number, in agreement with the naming e.g., (Wendisch et al., 2016)). The region of cloud base measurements is indicated by circles for each flight. The measurement strategy was developed such that measurements were made within at most 10 minutes and 60 km from each other. This was performed to assure that droplet measurements at cloud base pertain to the same air mass as the aerosol measurements below cloud base. A conceptual representation of the cloud profiling, including flight legs below and within cloud base, is shown for measurements during flight ~~AC19, moderate concentration during flights~~ *AC19 in Figure 1b. For the present study, the flight legs below and at cloud base are of primary relevance. Such flight legs, during which the relevant aerosol and cloud microphysical data were obtained, are distinguished by different colors. During flight AC19 the profiling of the marine shallow cumulus clouds was conducted up to an altitude of 4.3 km; details on the air mass origin and the aerosol properties during this flight can be found in Section 4.2. Aerosol properties were investigated during the flight leg below cloud base, which had a length of about 19 km at an altitude of ∼ 450 m above sea level (asl). Cloud microphysical properties of the marine shallow cumulus clouds (Atlantic ocean) were investigated during the flight leg near cloud base, which had a length of ∼ 60 km at an altitude at ∼ 600 m asl. A similar strategy was applied for in-land flights. Convective cumuli formed in very polluted environments (arc of deforestation) directly above the Amazonian deforestation arc during flight AC07. Less polluted clouds were found farther away from the deforestation fires over the tropical rain forest (remote Amazon) during flights AC09 and AC18.* ~~*and highest during flight AC07 in the southern and northern region of the Amazon Basin (Fig. S1)*~~

2.1 Aerosol size distribution below cloud base

Aerosol size distributions were measured using an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS; Droplet Measurement Technologies, Inc., Longmont, CO, USA) (Cai et al., 2008; Moore et al., 2021). The UHSAS combines a high-power infrared laser (λ = 1054 nm) and a large solid angle range in a side-ways direction for the detection of light scattered by individual particles (Andreae et al., 2018). The aircraft instrument measures particles in the diameter size range between 100 nm and 600 nm. The instrument is mounted in an under-wing canister. The sampled air is entering the instrument by a forward facing diffusor inlet, and the airflow is reduced by a second inlet to approximately isokinetic conditions. The measured particle diameter can be assumed to be close to their dry diameters due to heating effects (Chubb et al., 2016). The UHSAS was calibrated with monodisperse polystyrene latex (PSL) spheres of known size. Typical uncertainties of UHSAS measurements are both 15 % in diameter and concentration (Cai et al., 2008; Moore et al., 2021).

The total particle number concentration in the size range of ∼ 10 nm to ∼ 500 nm (N_{CN}) 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). N_{CN} was measured by a butanol-based condensation particle counter (CPCs, modified Grimm CPC 5.410 by Grimm Aerosol Technik, Ainring, Germany) with a flow of 0.6 L min^{-1}. Particle losses in the sampling lines have been estimated and taken into account with the particle loss calculator by von der Weiden et al. (2009). Typical uncertainties of CPC measurements are on the order of ∼10 % (Petzold et al., 2011).

The geometric mean of the aerosol size distribution and N_{CN} below cloud were calculated. The mean aerosol size distribution was fitted by one modal lognormal distributions. The integral of the fit for the aerosol size distribution should be similar to N_{CN}

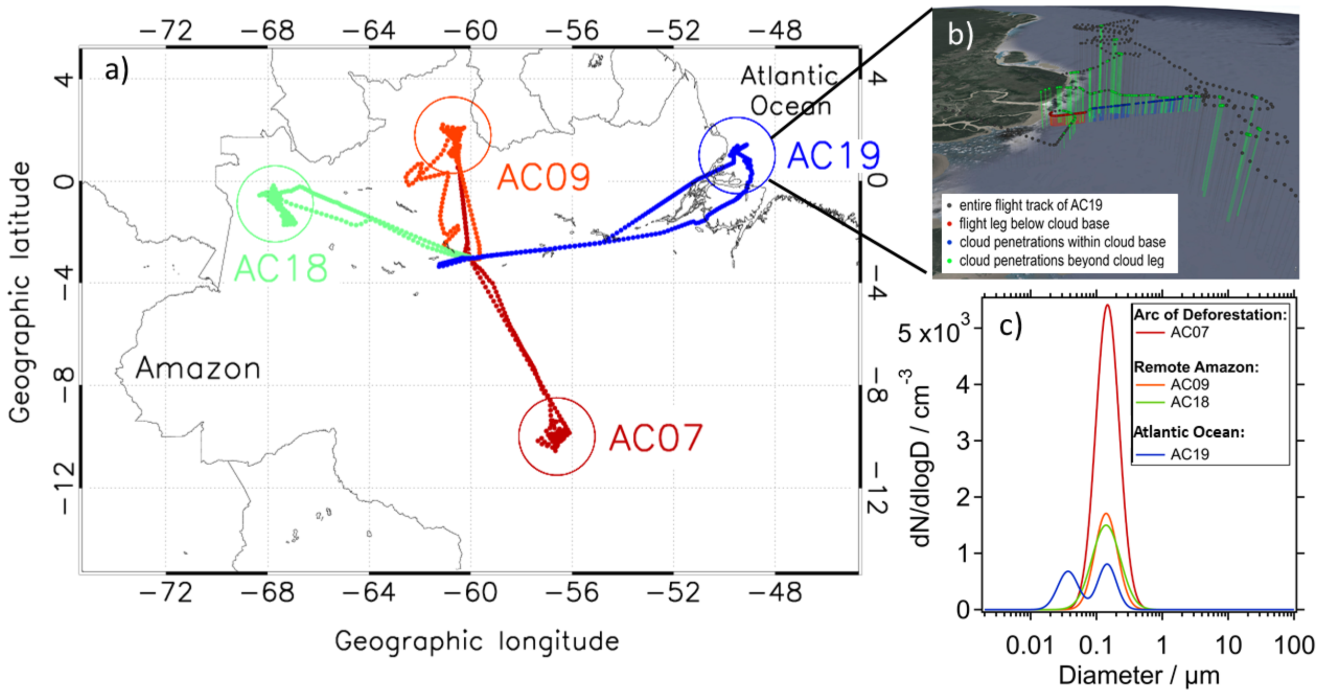

Figure R1-1. a) HALO flight tracks during the ACRIDICON–CHUVA experiment. The flight numbers are shown in the map in different colors. Colored circles indicate the region of aerosol and cloud measurements in each flight. The average aerosol particle concentration measured below cloud bases during flights AC07, AC09, AC18, and AC19 were 2417 cm^{-3}, 737 cm^{-3}, 809 cm^{-3} and 428 cm^{-3}, respectively. b) Cloud profiling maneuvers during flight AC19 above the Atlantic Ocean near the Amazon River delta shown as three-dimensional **(a)** profiles corresponding to the two dimensional profile (blue) in panel a). Relevant flight segments - particularly legs below cloud base and within cloud base, as well as cloud penetrations above cloud base - are emphasized by color-coding. c) aerosol size distributions for each flight as used in this study.

if mainly accumulation mode particles are present. This was fulfilled for AC07, AC09 and

The geometric mean of the aerosol size distribution and N_{CN} below cloud were calculated. The mean aerosol size distribution was fitted by a one modal lognormal distributions. The integral of the fit for the aerosol size distribution should be similar to N_{CN} if mainly accumulation mode particles are present. This was fulfilled for AC07, AC09 and ~~highest during flight AC07 in the southern and northern region of the Amazon Basin (Fig. S1;~~ AC18 but not for AC19 (Tables S1 - S4). ~~Figure 1 shows the measurement region for the textcolorBrickRedflights analysed in this study~~. For this latter flight, the integrated number concentration of the monomodal lognormal fit made up approximately half of the total N_{CN}. This discrepancy led to the assumption that a significant number concentration of particles in the size range of Aitken mode particles were present during AC19, but not captured by the UHSAS measurements. Consequently, a bimodal ASD shape was inferred. The geometric parameters for the lognormal distribution assumed for measurements during Flight AC19 were based on averages of bimodal aerosol size *distributions measured above the ocean in previous studies (Figure S4) (Wex et al., 2016; Quinn et al., 2017; Gong et al., 2019). The resulting shape of the two modes based on literature data was weighted by the difference between UHSAS and CPC measurements (Table S4). The number concentrations of all fitted aerosol size distributions were normalized to the measured N_{CN}. The variability of the aerosol number size distributions was calculated by the standard deviation on average ∼ 10 % and up to ∼ 20 % for very clean conditions. As a conservative approach ∼ 20 % was used in our model sensitivity study to take into account the impact of this variability on cloud droplet number concentration (Section 4.2). All concentrations are reported for normalized atmospheric conditions (Corrected for standard conditions (STP): T = 273.15°C and p = 1013.25 mbar).*

2.2 Cloud Droplet measurements at cloud base

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, Voigt et al., 2017). Cloud droplet number size distributions (DSDs) between 3 μm and 50 μm 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,Kleine et al., 2018,Wendisch and Brenguier, 2013). ~~Each DSD spectrum represents~~ These probes have different measurement characteristics such as particle inlet, sampling area of detection, size sensitivities etc. The CCP-CDP is an open-path instrument that detects forward-scattered laser light from cloud particles as they pass through the CDP detection area (Lance et al., 2010). CAS-DPOL collects forward-scattered light to determine particle size and number that pass the sampling area centered in an inlet shaft that guides the airflow. CCP-CDP and CAS-DPOL has similar values of uncertainty (∼ 10%) in the sample area. However, particle velocities in the sampling tube may be modified by the CAS tube when compared to the open path instruments (like CCP-CDP). This results in an additional uncertainty in the droplet number concentration measured by CAS-DPOL. During the ACRIDICON-CHUVA campaign the resulting uncertainty in the droplet concentration measured by CCP-CDP and CAS-DPOL were ∼ 10% and ∼ 21 %, respectively (Braga et al., 2017a)..

For cloud base measurements each probe DSD spectrum represented 1 s of flight path (covering between 70 m to 120 m of horizontal distance for the aircraft speed at cloud bases). We refer in the current study to the measurements closest to cloud base as 'cloud base' measurements, even if the actual cloud base might have been slightly below this altitude of measurements (Section 3.2.2 and Figure 2). 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 ± ∼ 30%~~ their uncertainty range for probe DSDs ($\pm \sim 16\%$). The overall systematic errors in the cloud probe integrated water content with respect to ~~$N_{d,m}$ at cloud bases~~ a King type hot-wire device are ∼ 6 % for CAS-DPOL and ∼ 21 % for CCP-CDP. A positive bias of ∼20% was found for ~~averaged~~ CAS-DPOL ~~measurements of $N_{d,m}$ at cloud bases~~ droplet concentration in comparison with those measured with CCP-CDP for cloud passes with cloud droplet effective radius < 7 μm (mostly measured at cloud bases). Cloud passes were defined for conditions under which the number droplet concentration (i.e., particles with diameter larger than 3 μm) exceeded 20 cm^{-3}. This ~~categorization~~ criterion was applied to avoid cloud passes well mixed with ~~environment air~~ subsaturated environment air (RH < 100%) and counts of haze particles. ~~The HALO aircraft was equipped with a meteorological sensor system (BAsic HALO Measurement And~~ Sensor System; BAHAMAS) ~~Updraft speeds at cloud base were measured in a range of 0.1 m s^{-1} located at the nose of the aircraft.~~typically found at cloud edges. Additional details about the cloud probes measurements at cloud bases used in this study can be found in Tables S1 and S2.

3. Methodology

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

The thermal instability in the boundary layer promotes the formation of clouds consisting of regions with updrafts and downdrafts. At cloud bases, the variability in vertical velocities and droplet concentration is high due to air turbulence. Clouds develop in updrafts, and during their vertical development the continued movement as a turbulent eddy adds a large random component to the relationship of w ~~$\leq$ 6 m s^{-1}, using a boom-mounted Rosemount model 858 AJ air velocity probe. The uncertainties in measured~~ with $N_{d,m}$. These intrinsic characteristics of clouds reduce the confidence that a measured w ~~are Δw < 0.2 m s^{-1} for~~ in the cloud led to the simultaneously measured $N_{d,m}$. Such inconsistencies often result in poor correlations of w and $N_{d,m}$.

~~The particle number concentration with diameter > 20 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 ~ 500 cm^{-3} to ~ 2000 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 Cai et al. (2008); Cai et al. (2011) with an estimated uncertainty of ~ 20% Andreae and Rosenfeld (2008). 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. he uncertainty of these size distribution measurements is estimated to be ±30%. These aerosol size distributions were used as input to the adiabatic parcel model (Section 3.2). 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.~~

In line XX, we modified the text as follows (l. 152):

Only cloud passes with positive w(i.e. updrafts) were considered in the analysis. Furthermore, we take into account only data of non-precipitating clouds, typically from cumulus humilis and cumulus mediocris clouds. Braga et al., 2017, have shown that ~~this method~~ the PMM can be used to find the best agreement between measured and estimated N_d at cloud base as a function of w.

In addition, we included two tables in the supplement (Tables S5 and S6 in the revised manuscript)

2 Referee comment: While the manuscript presents data from a valuable dataset, the analysis is not thorough, substantial amounts of detailed about the case study are missing and the conclusions are weak, and not evidence based. Based on my assessment, I suggest rejecting the manuscript.

Author response: In the revised manuscript version, we made clearer how we constrain the measurements on board the HALO aircraft to investigate the relationship between w, N_a, N_d and κ using airborne and model simulations. Please see our response to the previous comment and our responses to the specific comments below and updates throughout the text and supplement.

Table 1. (Table S5 in revised manuscript) Cloud probe size intervals and central bin diameters during HALO flights.

Cloud Probe	Size interval	Number of bins	Central bin diameter (μm)
CCP-CDP	3-50 μm	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
CAS-DPOL	3-50 μm	10	3.9, 6, 10.8 ,17.3, 22.3, 27.4, 32.4, 37.4, 42.4, 47.4

Table 2. (Table S6 in revised manuscript) Cloud base thermodynamic parameters and classification of each flight.

Flight	Classification	Altitude [m asl.]	Air Temperature [°C]	Time frame of measurements (UTC)]
AC07	Arc of Deforestation	1920	15	18:02:13 to 18:14:13
AC09	Remote Amazon	1200	19.5	15:40:41 to 15:58:35
AC18	Remote Amazon	1700	17	16:45:29 to 16:55:46
AC19	Atlantic Ocean	605	22	17:28:04 to 17:37:39

Specific comments:

3 Referee comment: Line 69 - please define AC. It would be more useful for the reader to name the cases by their attributes than their flight number. Something like LPC- low particle concentration, MCP1,MCP2 –moderate particle concentration...etc.
Author response: The flights are label with AC and a running number, we did make this clear in the manuscript. We prefer to continue using these labels in our study for consistency with previous studies on the ACRIDICON campaign in which the same flight names were used, e.g., (Braga et al., 2017; Cecchini et al., 2017; Holanda et al., 2020; Wendisch et al., 2016). However, we added a description of the condition to the flight number.
We added to the manuscript (l. 67):
Figure 1a shows the measurement region for the flights analyzed in this study (the flights are labelled with 'AC' and a running number, in agreement with the naming e.g., (Wendisch et al., 2016). We added a new Table S6, that also provides a characterization of the air masses)

4 Referee comment: Line 71- What do you mean by environment air? subsaturated air?
Author response: We revised the text as follows (l. 136):
This categorization was applied to avoid cloud passes well mixed with subsaturated environment air (RH < 100%) and counts of haze particles, typically found at cloud edges.

5 Referee comment: Line 73- By "at cloud base" do you actually mean at cloud base or was it slightly above or below cloud base?

Author response: We assume that measurements was taken slightly above cloud base. To investigate at which height above cloud base measurements was performed the measured liquid water content (LWC) was compared to the modeled LWC and the best match was found at 20 m above cloud base as shown in Figure 2. We made this more clear by adding the following sentence into the manuscript (l. 128):

We refer in the current study to the measurements closest to cloud base as 'cloud base' measurements, even if the actual cloud base might have been slightly below this altitude of measurements (Section 3.2.2 and Figure 2).

6 Referee comment: Line 78 – where were these size distribution measurements made relative to the cloud measurments? Were they averaged before fitted to log normal distributions?

Author response: the measurement strategy was developed such that measurements were made within at most 10 minutes and 60 km from each other, respectively. This was performed to assure that droplet measurements at cloud base pertain to the same air mass as the aerosol measurements below cloud base. We made this more clear by adding a map showing the flight segment below and at cloud base in Figure 1 and adding the following statement to the manuscript (l. 69):

The measurement strategy was developed such that measurements were made within at most 10 minutes and 60 km from each other. This was performed to assure that droplet measurements at cloud base pertain to the same air mass as the aerosol measurements below cloud base.

The aerosol size distributions was averaged first and after this the fitting was performed. This is well described in the revised Methodology section (l. 98):

The geometric mean of the aerosol size distribution and N_{CN} below cloud were calculated. The mean aerosol size distribution was fitted by one modal lognormal distributions.

7 Referee comment: Line 80 – Please provide more detail on how the Aitken mode was inferred. It is unclear since the UHSAS only measured as low as 60 nm.

Author response: We added the following clarification to the text (l. 98ff).

The geometric mean of the aerosol size distribution and N_{CN} below cloud were calculated. The mean aerosol size distribution was fitted by one modal lognormal distributions. The integral of the fit for the aerosol size distribution should be similar to N_{CN} if mainly accumulation mode particles are present. This was fulfilled for AC07, AC09 and ~~and highest during flight AC07 in the southern and northern region of the Amazon Basin (DFig. S1;~~ AC18, but not for AC19 (Tables S1-S4). ~~Figure 1 shows the measurement region for the flights analyzed in this study.~~ For this latter flight, the integrated number concentration of the monomodal lognormal fit made up approximately half of the total N_{CN}. This discrepancy led to the assumption that a significant number concentration of particles in the size range of Aitken mode particles were present during AC19, but not captured by the UHSAS measurements. Consequently, a bimodal ASD shape was inferred. The geometric parameters for the lognormal distribution assumed for measurements during Flight AC19 were based on averages of bimodal aerosol size distributions measured above the ocean in previous studies (Figure S4) (Wex et al., 2016; Quinn et al., 2017; Gong et al., 2019). The resulting shape of the two modes based on literature data was weighted by the difference between UHSAS and CPC measurements (Table S4). The number concentrations of all fitted aerosol size distributions were normalized to the measured N_{CN}. The variability of the *aerosol number size distributions was calculated by the standard deviation on average ∼ 10 % and up to ∼ 20 % for very clean conditions. As a conservative approach ∼ 20 % was used in our model sensitivity study to take into account the impact of this variability on cloud droplet number concentration (Section 4.2). All concentrations are reported for normalized atmospheric conditions (Corrected for standard conditions (STP): T = 273.15°C and p = 1013.25 mbar).*

8 Referee comment: Line 91 – did you demean the updraft velocity? What do you mean by passes with only positive w were considered? Surely there were often some Negative values? I am guessing you mean you just excluded negative values?

Author response: We added the following information in the manuscript.

line 142: *The thermal instability in the boundary layer promotes the formation of clouds consisting of regions with updrafts and downdrafts. At cloud bases, the variability in vertical velocities and droplet concentration is high due to air turbulence. Clouds develop in updrafts, and during their vertical development the continued movement as a turbulent eddy adds a large random component to the relationship of w with $N_{d,m}$.* ~~*$w<5$ m s^{-1} and $\Delta w \approx 0.25$ m s^{-1} for $w>5$ m s^{-1}. Further details on the uncertainties of w measurements are described by Mallaun et al. (2015)*~~

line 145. *These intrinsic characteristics of clouds reduce the confidence that a measured w in the cloud led to the simultaneously measured $N_{d,m}$. Such inconsistencies often result in poor correlations of w and $N_{d,m}$.*

line 152 *Only cloud passes with positive w (i.e. updrafts) were considered in the analysis. Furthermore, we take into account only data of non-precipitating clouds, typically from cumulus humilis and cumulus mediocris clouds.*

9 Referee comment: Line 106 – Was there any drizzle in the measured clouds? It makes sense to exclude collision coalescence in a parcel model since it cannot be parameterized will with a 0D model, however can you confirm with measurements and results from previous modeling studies that Collision/coalescence are negligible in the clouds you studied (at least the lower 70 m).

Author response: No, we did not have drizzle or rain at cloud base; only non-precipitating clouds were considered (cf our response to previous comment). This was checked by CIPgs. We added also more information regarding previous model studies of collision/coalescence (l. 168)

Collision/coalescence processes are not considered as we restrict our analysis to heights near cloud base where droplets are relatively small and the cloud droplet size distribution is narrow. Under such conditions, collision-coalescence is likely negligible (Rosenfeld, 2018; Shaw et al., 1998; Xue et al., 2008).

10 Referee comment: Line 112 - "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 k= 0.1, together with w measured at cloud base." It is not clear how this I used to determine the height at which $N_{d,m}$ and $N_{d,p}$ should be compared.

Author response: We have improved the text (l. 176ff):

In order to determine the height at which $N_{d,m}$ and $N_{d,p}$ should be compared, the measured liqued water contend (LWC) was compared to the simulated LWC using the aerosol size distribution for the different flights together with w measured at cloud base and assumed hygroscopicity of $\kappa = 0.1$. ~~*simulations were performed using the measured aerosol particle size distributions*~~

~~*and an assumed hygroscopicity of κ = 0.1, together with w measured at cloud base.*~~ *Under adiabatic conditions, $N_{d,p}$ is predicted to be approximately constant at ∼ 20 m above the level of the maximum supersaturation S_{max} (Fig. S6). Figure 2 shows the frequency of measured LWC and the modeled LWC at different heights. At ∼ 20 m above cloud base the LWC measured with the highest frequency and the modeled LWC is the same. For this reasoned the model results at 20 m above cloud base are compared to the measured cloud droplet number concentrations in the scope of this study.*

11 Referee comment: Line 114- make it clear that this is a result from your adiabatic parcel model, not measurements.
Author response: We revised the sentence as follows (l. 178):
Under adiabatic conditions, $N_{d,p}$ is predicted to be approximately constant at 20 m above the level of the maximum supersaturation Smax (Fig.S3).

12 Referee comment: Line 120 "in the following..." Section?
Author response: Yes, we added 'Section' (now: Line 178).

13 Referee comment: Line 133 – how do you define best agreement? Absolute concentration? Percentage difference?
Author response: The simulated values for droplet concentration that have the best agreement with measurements as a function of κ and w are defined based on smaller bias, root mean square and standard error. We added the following tables in the supplement showing these statistics (absolute bias, RMSE, mean absolute error and ratio, new Tables S7 – S18 in the supplement).

14 Referee comment: Line 145 – For lateral entrainment to increase droplet concentration, the entrained concentration would need to be both CCN active and higher than the below cloud concentration. I have never seen a case where this has occurred. Can you cite a relevant source? Also, even if there was lateral entrainment, at altitudes so close to cloud base, it is unlikely that there is a significant influence from lateral entrainment. Entrainment would also likely dry the air, decreasing the supersaturation, further decreasing the chance of increasing the droplet concentration.
Author response: We agree with the referee that it seems unlikely that entrainment leads frequently to increased particle and therefore droplet concentrations. We weakened this statement and also removed at other places any text about entrainment (l. 224)
~~*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)*~~. *However, while entrainment of biomass burning aerosol may be possible, we do not have any quantitative information on such processes.*

15 Referee comment: Line 147 – Entrainment typically leads to a decrease in N_d.
Author response: We agree with the referee; please see our response to Comment 14.

16 Referee comment: Line 150 – why is that unlikely?
Author response: This is now explained in more detail in the revised manuscript (l. 226):
While also particles of different hygroscopicities and activation thresholds depending on w might explain the trends in Fig.

3a-c, there is no indication of higher hygroscopicity of smaller accumulation mode aerosol particles during the Amazonian dry season (e.g., Pöhlker et al., 2016, 2018). In air masses of different origin, aerosol particles would likely not only exhibit different chemical composition and hygroscopicity but also large variability in their particle number concentrations. Given the relatively small standard deviations in the measured N_a (Tables S1 - S4), we are confident that the sampled aerosol populations did not have large variability in their composition.

We have extended Tables S1-S4 with additional information. We also report now all concentrations for normalized atmospheric conditions (STP corrected to T = 273.15°C and p = 1013.25 mbar).

Tables S1 - S4: Geometric mean and standard deviation of atmospheric parameters measured below cloud base. Note that the mean diameter is a fit parameter of the average aerosol size distribution and the error is 15 % according to (Cai et al., 2008; Moore et al., 2021).

Flight AC07

6-09-2014 17:53:00 to 17:55:25 UTC

Parameter	Mean ± SD
Altitude [m asl.]	1800 ± 0.05
Air Temperature [°C]	18 ± 0.15
Pressure [hPa]	820 ± 0.25
Relative Humidity [%]	95 ± 3
N_{CN} [cm^{-3}]	2417 ± 42
N_{UHSAS} [cm^{-3}]	2024 ± 162
d_{acc} [nm]	147 ± 22

Flight AC09

11-09-2014 15:31:31 to 15:38:20 UTC

Parameter	Mean ± SD
Altitude [m asl.]	933 ± 0.05
Air Temperature [°C]	22.5 ± 0.15
Pressure [hPa]	938 ± 0.25
Relative Humidity [%]	87 ± 3
N_{CN} [cm^{-3}]	737 ± 58
N_{UHSAS} [cm^{-3}]	686 ± 59
d_{acc} [nm]	140 ± 22

Flight AC18

28-09-2014 16:39:00 to 16:43:59 UTC

Parameter	Mean ± SD
Altitude [m asl.]	1286 ± 0.05
Air Temperature [°C]	20.7 ± 0.15
Pressure [hPa]	876 ± 0.25
Relative Humidity [%]	81.8 ± 2
N_{CN} [cm^{-3}]	809 ± 20
N_{UHSAS} [cm^{-3}]	707 ± 83
d_{acc} [nm]	140 ± 21

Flight AC19

30-09-2014 17:23:38 to 17:27:31 UTC

Parameter	Mean ± SD
Altitude [m asl.]	452 ± 0.05
Air Temperature [°C]	23.5 ± 0.15
Pressure [hPa]	960 ± 0.25
Relative Humidity [%]	93.7 ± 3
N_{CN} [cm^{-3}]	428 ± 138
N_{UHSAS} [cm^{-3}]	227 ± 52
N_{ait} [cm^{-3}]	∼ 201
N_{acc} [cm^{-3}]	227 ± 52
d_{ait} [nm]	∼ 37
d_{acc} [nm]	136 ± 20

17 Referee comment: Line 155- What evidence is there for this cloud being impacted by marine air? Having a bimodal distribution does not make an aerosol distribution impacted by marine air. It is very likely that all of the particle distributions have a bimodal distribution, however you cannot tell because you are limited to a minimum particle size measurement of 60 nm by the UHSAS. It is still unclear how you obtained an Aitken mode for AC19.

Author response: We agree with the referee that bimodality does not necessarily imply that the air mass had marine origin and vice versa. As it was misleading in the text, we changed it as follows (l. 238):

The air masses below cloud encountered during flight AC19 were mostly impacted by marine air ~~leading to~~ (as supported by prior back trajectory analysis (Section S1 and Holanda et al. (2020)) and exhibited a bi-modal aerosol size distribution with low $N_{d,m}$ (Figure 1c).

We had strong indications that indeed a monomodal aerosol distribution was not sufficient to explain the observed N_a. We added more details on the fitting procedure of the bimodal size distributions (see our response to Referee Comment 7).

18 Referee comment: Line 157 – this result suggests that the aerosol you measured and the aerosol that entered the cloud are not from the same population.

Author response: We added more details on the aerosol and cloud measurements in Section 2. (See our responses to your general comments 1 and 2 at the beginning of this response). In addition, we also added more explanation on the reasons why the N_d closure is worse for AC19 than for the other flights. In brief, we argue that likely the assumption of identical κ values for both modes is an oversimplification and that properties for Aitken and accumulation modes need to be taken into account.

19 Referee comment: Line 165 The following two quotes from your text are inconsistent with your argument "The chemical composition of Aitken mode particles often differs significantly from that of accumulation mode particles, which are more aged and internally mixed" "Marine particles often show similar hygroscopicity in both Aitken and accumulation modes"

Author response: We agree with the referee that the text was contradictory as it was written. We removed the respective sentence: ~~*Marine particles often show similar hygroscopicity in both Aitken and accumulation modes (Kristensen et al., 2016; Wex et al., 2016).*~~

20 Referee comment: Line 168 – You have indicated it is possible that the hygroscopicity of these particles may be inconsistent with the values you used because you think they are marine aerosol. You have the ability to test this hypothesis with your parcel model, but choose not to. Why?

Author response: We thank referee for this suggestion and agree that further sensitivity simulations added value to our study. We performed additional studies using all combinations of κ = 0.1 and 0.6 (and 0.8/0.1) for the two modes. Results are shown in Figure 3 e and discussed as follows (l. 253ff):

To account for different hygroscopicities in Aitken and accumulation modes, we performed further sensitivity analyses using combinations of κ = 0.1 and 0.6 for the two modes (Figure R1-2e). It is obvious that the choice of κ for the Aitken mode (κ_{Ait}) does not affect $N_{d,p}$ for $w \leq\sim 1$ m s^{-1} in the presence of very hygroscopic accumulation mode particles (κ_{acc} = 0.6) or below $w \leq\sim 0.5$ m s^{-1} with κ_{acc} = 0.1, respectively. Even assuming rather extreme values of κ_{Ait} = 0.8 cannot fully reproduce the large increase in N_d at $w \geq\sim 1.5$ m s^{-1} as observed by the CAS probes; assuming very hygroscopic Aitken mode and less hygroscopic accumulation mode particles can approximately reproduce the trend in $N_{d,m}$ from the CDP.

Varying κ_{acc} from 0.1 to 0.6 leads to a large increase of $N_{d,p}$ at all w. The corresponding change in $N_{d,p}$ by increasing κ_{Ait} is much smaller. The reason for this relatively smaller sensitivity of $N_{d,p}$ to κ_{Ait} is the fact that the supersaturation in the cloud is mostly controlled by the droplet growth on accumulation mode particles. The sensitivity of $N_{d,p}$ formed on Aitken mode particles to κ_{acc} is slightly larger if κ_{acc} = 0.1 as compared to κ_{acc} = 0.6 because in the latter case, the supersaturation is efficiently suppressed preventing a higher number of Aitken mode particles from activating. Overall we can conclude that assuming different κ values for accumulation and Aitken mode leads to a better representation of the observed trends of $N_{d,m}$ with w (Tables S16 ad S17). However, in the absence of more information on the particle hygroscopicity we cannot state with certainty that indeed the assumptions of the two κ values are appropriate for this aerosol population. 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 the particle number concentrations of the accumulation and Aitken modes are comparable. By using a single κ value, we cannot reproduce the observed continuously strong increase of $N_{d,m}$ for the whole w range. Instead we predict a smaller increase at w $\sim$ 1 m s^{-1}, i.e., a flattening of the curve.

Figure 3 was accordingly updated. Please see Figure R1-2. As indeed the simulations assuming κ_{Ait} = 0.6 and κ_{acc} = 0.1 or κ_{Ait} = 0.8 and κ_{acc} = 0.2 give much better results, we also discuss them now in the text of the abstract and summary and conclusion section:

Abstract: *Above the ocean, fair agreement was obtained assuming an average hygroscopicity of $\kappa \sim 0.2$ (deviations $\leq\sim$ 16%) and further improvement was achieved assuming different hygroscopicities for* Aitken and accumulation *mode particles (κ_{Ait} = 0.8, κ_{acc} = 0.2; deviations $\leq\sim$ 10%), which may reflect secondary marine sulfate particles. Our results indicate* that Aitken mode particles *and their hygroscopicity can be important for droplet formation at low pollution levels and high updraft velocities in tropical convective clouds.*

Summary and conclusions*: Above the western Atlantic best N_d closure was achieved for $\kappa \sim 0.2$ applying a single κ value for both Aitken and accumulation modes; an even better representation of the increase in N_d with w was obtained when moderately hygroscopic accumulation mode particles (κ_{acc} = 0.2) and highly hygroscopic Aitken mode particles (κ_{Ait} = 0.8) were assumed.*

21 Referee comment: Line 172- you should include sensitivity calculations.

Author response: We added the new Figure S7 to the supplement. In addition, we added some detail and discussion about the calculation and interpretation of the sensitivities in the new Section 4.3 (Please our detailed response to Referee Comment 28.)

22 Referee comment: Line 175- do you mean additional aerosol? Additional activation would surely lead to additional activated particles.

Author response: We agree with the referee that the sentence was misleading. We reworded it as follows (l. 407):

In these studies, it was demonstrated that ~~at high w, the activated particle fraction is sufficiently high that additional activation does not lead to a significant increase in activated particles, and, thus, the sensitivity to~~ the sensitivity of N_d to becomes small. ~~In these studies~~ Our analysis shows that measurement uncertainties in basic aerosol properties might equally explain such differences. If particles exceed a hygroscopicity threshold ($\kappa >\sim 0.3$), predicted cloud droplet number concentration becomes very insensitive to κ when a large fraction of all particles are activated ('aerosol-limited regime').

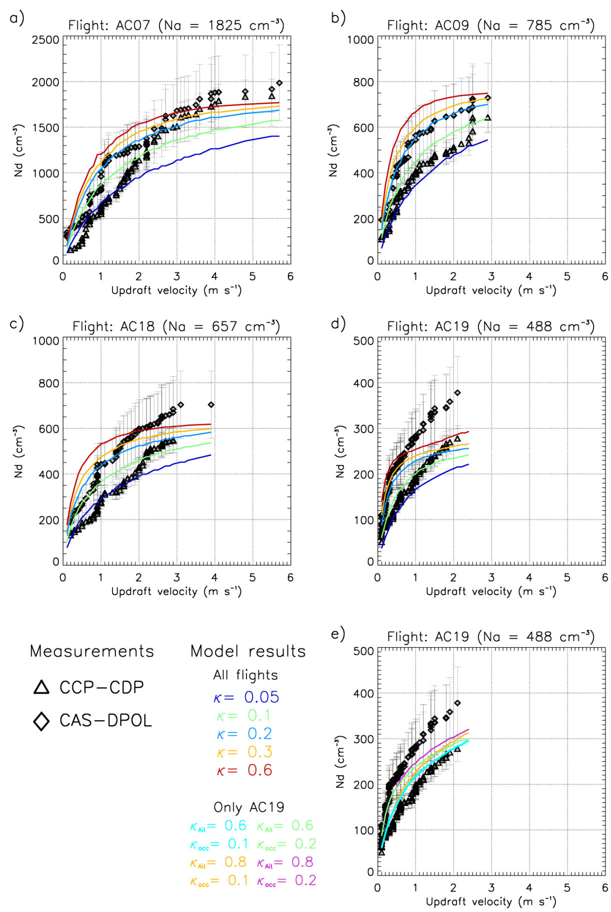

Figure R1-2. (= Figure 3 in the revised manuscript) Cloud droplet number concentration (N_d) as a function of updraft velocity near cloud base of convective clouds during flights: a) AC07, b) AC09, c) AC18, d) and e) AC19. The measured updraft velocities are based on the "probability matching method" (PMM) using the same percentiles for updraft velocity and $N_{d,m}$ (Section 3.1). The black diamond and triangle symbols represent $N_{d,m}$ near cloud base with the CAS-DPOL and CCP-CDP probes, respectively. Measurement uncertainties (indicated by error bars) are $\sim$ 21% and $\sim$ 10% for CAS-DPOL and CCP-CDP data (Braga et al. (2017a)). The colored lines in panels a)-d) show $N_{d,p}$ assuming a single κ value for both modes (labeled on the left). Panel e) shows $N_{d,p}$ based on simulations assuming different values of κ for Aitken and accumulation mode particles.

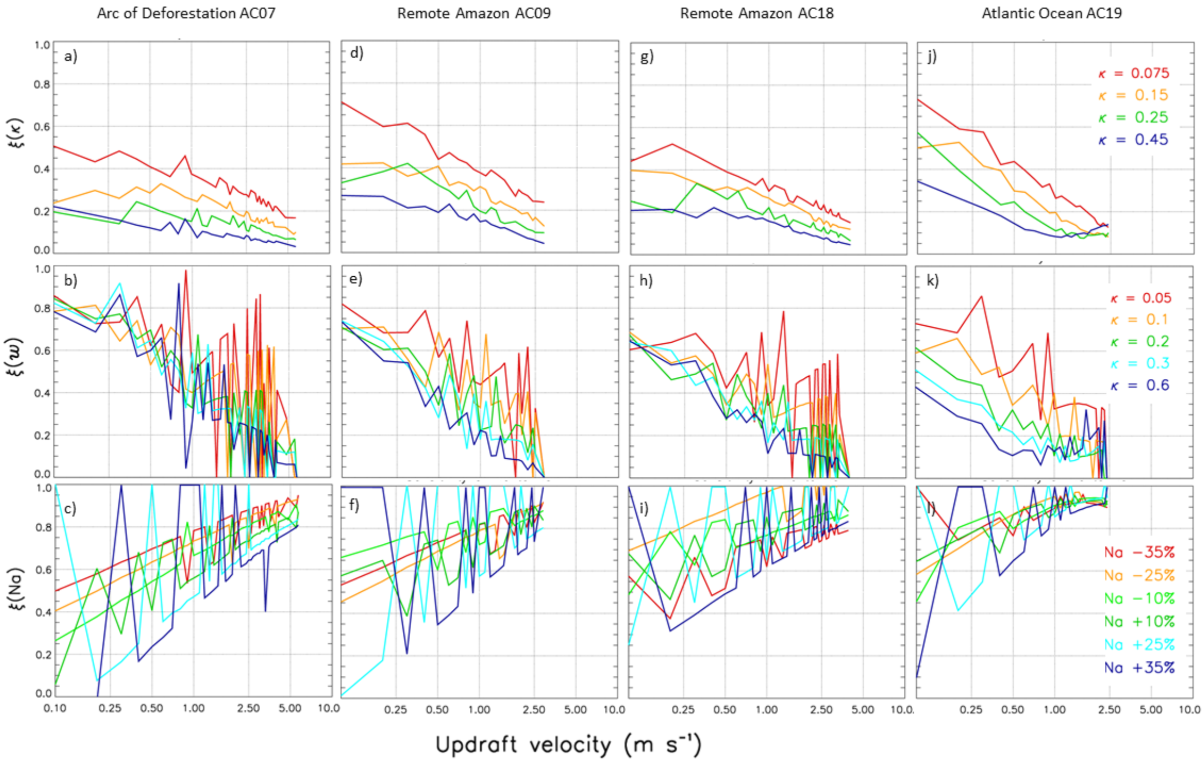

Figure R1-3. (= Figure S7 in the revised manuscript) Modeled sensitivity of droplet number concentration (N_d) to changes in the hygroscopicity parameter κ, vertical velocity (w) and aerosol number concentration (N_c) for the measured conditions during flight AC07 (arc of deforestation), AC09 and AC18 (remote Amazon) and AC19 (Atlantic ocean).

23 Referee comment: Line 178 – "high sensitivities of N_d to the chemical composition of Aitken mode particles might affect cloud properties" This statement is redundant.

Author response: We reworded the sentence as follows (l. 277):

~~*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.*~~*Anttila and Kerminen (2007) showed in a model study focusing only on Aitken mode particles that N_d is highly sensitive to the chemical composition of Aitken mode particles.*

24 Referee comment: Line 179 – comparable in composition? Concentration? This is unclear. This statement about kappa contradicts your statement in line 167.

Author response: We are not sure which line the referee is referring to. As the 'comparable' in the following sentence may have led to the confusion, we clarified it as follows (l. 268):

when ~~N_a~~the particle number concentrations of the accumulation and Aitken modes are comparable.

With the regard to the statement about κ in the two modes, we added some findings from our most recent study (Pöhlker et al., 2021) that was published in ACPD nearly concurrently with the ACPD version of the present manuscript. There we show that the sensitivities of κ and N_a to N_d are different for accumulation and Aitken mode particles, respectively.

Therefore, our statements are not contradicting each other as in the previous text, we referred to accumulation mode particles. We clarify this now as follows (l. 278):

In our recent model study, we systematically explored the extent to which the presence of an Aitken mode might significantly affect N_d as a function of updraft velocity (Pöhlker et al., 2021). In that study, we show that the sensitivities of $N_{d,p}$ are different to the properties (N_a, κ) of accumulation and Aitken mode particles, respectively. Generally, we find that $N_{d,m}$ is not highly sensitive to Aitken mode particle properties in the presence of a dominant accumulation mode, which is in agreement to our results in Figures 3 and S7.

25 Referee comment: Line 186- how was this 30% uncertainty calculated?

The uncertainty is actually only 20%, being the sum of 10% uncertainty for aerosol size distribution measurements and 10% uncertainty in the measurement N_d. We noticed that the 30% mentioned in the previous manuscript version were too high as one of the uncertainty was erroneously double-counted.

We added the error bars for $N_{d,m}$ in the figures. To explore the sensitivities of $N_{d,p}$ to an even wider range of N_a, we included now model results for different N_a (±20 %, ±30 %, ±40 %) to not only cover the uncertainty and variability in measurements but also show the sensitivity of N_d to N_a. In addition we expend the discussion about uncertainty and variability of the measured aerosol size distribution. Accordingly, the following changes have been bade in the manuscript (l. 92ff):

The total particle number concentration in the size range of ∼ 10 nm to ∼ 500 nm (N_{CN}) 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). N_{CN} was measured by a butanol-based condensation particle counter (CPCs, modified Grimm CPC 5.410 by Grimm Aerosol Technik, Ainring, Germany) with a flow of 0.6 L min^{-1}. Particle losses in the sampling lines have been estimated and taken into account with the particle loss calculator by von der Weiden et al. (2009). Typical uncertainties of CPC measurements are on the order of ∼10 % (Petzold et al., 2011).

The geometric mean of the aerosol size distribution and N_{CN} below cloud were calculated. The mean aerosol size distribution was fitted by one modal lognormal distributions. The integral of the fit for the aerosol size distribution should be similar to N_{CN} if mainly accumulation mode particles are present. This was fulfilled for AC07, AC09 and AC18, but not for AC19 (Tables S1-S4). For this latter flight, the integrated number concentration of the monomodal lognormal fit made up approximately half of the total N_{CN}. This discrepancy led to the assumption that a significant number concentration of particles in the size range of Aitken mode particles were present during AC19, but not captured by the UHSAS measurements. Consequently, a bimodal ASD shape was inferred. The geometric parameters for the lognormal distribution assumed for measurements during Flight AC19 were based on averages of bimodal aerosol size distributions measured above the ocean in previous studies (Figure S4) (Wex et al., 2016; Quinn et al., 2017; Gong et al., 2019). The resulting shape of the two modes based on literature data was weighted by the difference between UHSAS and CPC measurements (Table S4). The number concentrations of all fitted aerosol size distributions were normalized to the measured N_{CN}. The variability of the aerosol number size distributions was calculated *by the standard deviation on average ∼ 10 % and up to ∼ 20 % for very clean conditions. As a conservative approach ∼ 20 % was used in our model sensitivity study to take into account the impact of this variability on cloud droplet number concentration (Section 4.2). All concentrations are reported for normalized atmospheric conditions (Corrected for standard conditions (STP): T = 273.15°C and p = 1013.25 mbar).*

Figure 4 was replaced by the following figure and the text was adjusted accordingly.

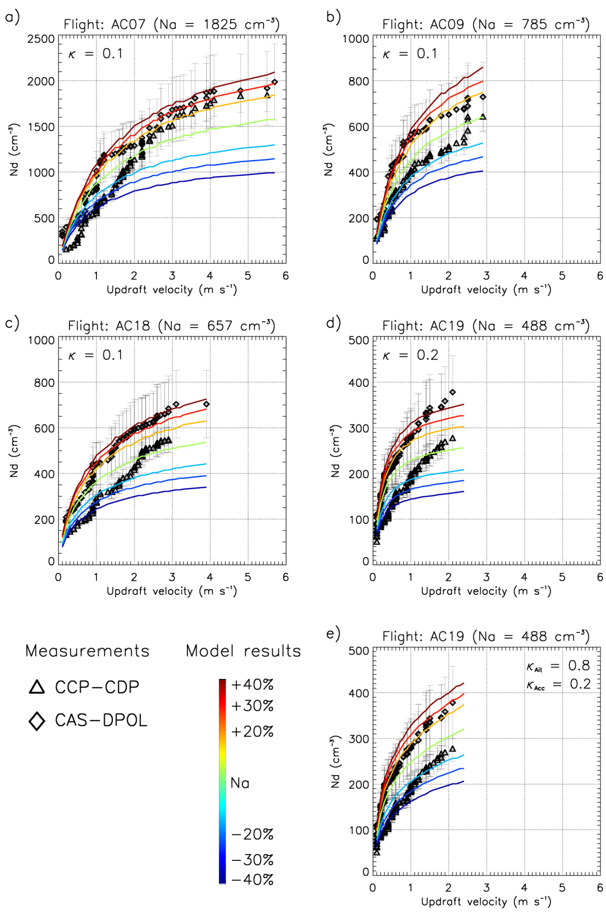

Figure R1-4. (= Figure 4 in the revised manuscript) Cloud droplet number concentration (N_d) as a function of updraft velocity near cloud base of convective clouds during flights: a) AC07, b) AC09, c) AC18, d) and e) AC19. The measured updraft velocities are based on the "probability matching method" (PMM) using the same percentiles for updraft velocity and $N_{d,m}$ (Section 3.1). The black diamond and triangle symbols represent $N_{d,m}$ near cloud base with the CAS-DPOL and CCP-CDP probes, respectively. Measurement uncertainties (indicated by error bars) are ∼ 21% and ∼ 10% for CAS-DPOL and CCP-CDP data (Braga et al. (2017a)). The lines show $N_{d,p}$ assuming the uncertainty range of N_a measurements, colored-coded by ΔN_a [%].

26 Referee comment: Line 215 – non-adiabatic conditions like entrainment would not increase particle concentration, only decrease.

Author response: We agree with the referee that the discussion of entrainment as an explanation for resulting higher N_d does not seem likely. Therefore, we weakened this statement as follows (l.394):

~~The~~ Our comparison between predicted and measured N_d showed largest discrepancies at high updraft velocities (w > 2.5 m s-1), which ~~may be~~ could be possibly explained by non-adiabaticity and/or entrainment of aerosol particles near cloud bases of convective clouds.

27 Referee comment: Line 217 calculations are necessary to back this claim and could easily be performed.

Author response: We agree with the referee that the predicted differences in N_d due to variation in κ or N_a could have been more quantified. Statistical parameters in Tables S7-S18 have been added and are discussed in Section 4.1 and 4.2. The new Section 4.3 also provides more information on such sensitivities in the present and previous studies.

28 Referee comment: Line 218: you should be able to state your conclusions based on the evidence provided in the manuscript, not by citing others work.

Author response: We agree with the referee that the placement of these references was not ideal. We moved them now to the new Section 4.3 (Sensitivities of N_d predictions to w, N_a and κ in the context of previous studies and restricted the text in the Summary and conclusions Section to conclusions based on our study.

4.3 Sensitivities of N_d predictions to w, N_a and κ

The sensitivities of cloud drop number concentrations to hygroscopicity (κ), N_a and w have been explored in numerous previous studies. In the following, we place our results in the context of such studies. Consistent with such previous sensitivity studies, we calculated the sensitivity ξ of $N_{d,p}$ to κ, N_a and w

$$\xi(X) = \frac{\partial ln N_d}{\partial ln X} \quad \text{(E.1)}$$

whereas X is κ, N_a or w, respectively. The results are summarized in Figure S7. They show that $\xi(\kappa)$ is smallest as compared to $\xi(N_a)$ and $\xi(w)$. $\xi(\kappa)$ is highest for low κ as conditions, the activated fraction is smallest and thus a small change in κ might cause a significant change in N_d. Generally, sensitivities are high under conditions of high supersaturation which are present at high w and/or low N_a.

While the sensitivities calculated for flights AC07, AC09 and AC18 follow these general trends according to N_a, the $\xi(X)$ values are higher for AC19 even though N_a was lowest for this flight. The reason for this difference is the successive activation of Aitken mode particles at high w Pöhlker et al. (2021). A sensitivity study for the same flights has been performed previously (Cecchini et al., 2017) in which the sensitivity of N_d and effective cloud droplet diameter to N_a and w was explored in detail at various heights in cloud. The focus of that study was the change of the sensitivities during cloud evolution, i.e. as a function of height in cloud. In the present study, we focus on the sensitivities of N_d near cloud base, but additionally explore the importance of κ in determining N_d. Such analysis can be used to give guidance for future measurements in similar clouds on the absolute values and relative importance of the three parameters to predict N_d.

Generally, prior sensitivity studies agree in the rankings of the relative importance of hygroscopicity (κ), N_a and w, as also shown in Figure S7. Feingold (2003) has shown that N_a has the largest influence on effective radius which is indirectly related to N_d. The sensitivities to the effective radius are typically smaller than those to N_d (Pardo et al., 2019). In our recent model study, we have shown that in the transitional regime, i.e., in the parameter space between the aerosol- and updraft limited *regimes, as defined by Reutter et al. (2009), N_d can be equally sensitive to κ and w (Pöhlker et al., 2021). In that study, we show that with increasing N_a, the sensitivities to both parameters decrease; however, the sensitivity of N_d to w remains higher under such conditions than that to κ.*

The uncertainties in updraft measurements are larger than those of hygroscopicity due to the great variability of w near cloud base. Peng et al. (2005) compared $N_{d,p}$ based on a w distribution in a range of 0.09 - 1 m s^{-1} and using characteristic single w values. They found differences in $N_{d,p}$ on the order of < 10% for the two sets of model simulations. Meskhidze et al. (2005) performed model simulations of low-level cumuliform clouds for which a range of 0.9 m $s^{-1} \leq w \leq 2.8$ m s^{-1} had been observed. They concluded that parameterizations of $N_{d,p}$ should include a weighting factor for high values of w as otherwise N_d might be biased high due to enhanced vertical velocity within in cloud cores as compared to cloud base.

In turbulent clouds with high w, the determination of w near cloud bases might be challenging; however, the resulting uncertainties in updraft velocity or its distributions cannot explain the discrepancies between $N_{d,m}$ and $N_{d,p}$ at high w (Figures 3 and 4). Under such conditions, the activated fraction approaches unity and any increase in w would not lead to higher N_d and improve the overall N_d closure [e.g., Hsieh et al. (2009)]. Therefore under such condition, ξ(w) becomes small (Figure S7b, e, h, k) These previous N_d sensitivity and closure studies either considered w as a fitting parameter to obtain good closure or used w values or distributions as relatively poorly constrained parameters. The PMM analysis as applied in the current study partially overcomes these uncertainties as it provides a stronger constraint of the w and N_d pairs for the full w range (Section 3.1), as opposed to the previous studies that derived their w distributions from averaging measured updraft velocities without sorting w and $N_{d,m}$ data based on their frequency occurrence.

Reutter et al. (2009) termed conditions under which nearly all particles are activated into cloud droplets as 'aerosol-limited regime' when N_d is only dependent on N_a, and not on w. Such conditions are present at relatively low total N_a and high w, i.e., when the maximum supersaturation in the cloud is relatively high. When an increase in N_a results in an equal increase in N_d, $\xi(N_a)$ approaches unity (Figure S7c, f, i, l). The measured and predicted activated fractions for flights AC07, AC09 and AC18 reach ≥80% at updraft velocities of $w \geq\sim 1$ m s^{-1} if the measured value is based on the CAS data (Figure 4). Therefore, we conclude that the sensitivity of N_d to N_a is much greater than that to w under these conditions which is also reflected by the rather small increase in N_d with w at high updraft velocities.

Overall, the variability of predicted N_d due to inferred κ ranges in the present study confirm trends from previous sensitivity studies for mono-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 (Figure S7 and Ervens et al. (2005); Reutter et al. (2009); Cecchini et al. (2017); Pardo et al. (2019)). If low hygroscopicity limits the water vapor uptake, a small change in κ may lead to a significant change in N_d, resulting in high $\xi(\kappa)$ values. A change of κ by the same factor for highly hygroscopic particles, however, might not lead to a significant change in N_d due to the regulation of the supersaturation ('buffering'), i.e., the efficient growth of more cloud droplets which, in turn, reduces the supersaturation. Our sensitivity study of AC19 exceeds these previous sensitivity studies that focused on monomodal aerosol size distributions. We show that the uncertainties in $N_{d,p}$ become larger under conditions when Aitken mode particles contribute to N_d as only at very high w the aerosol-limited regime is reached and $\xi(\kappa)$ and $\xi(w)$ decrease. Qualitatively this was also suggested in a previous N_d closure study for marine stratocumulus clouds, where it was concluded that only the presence of an Aitken mode could explain the high $N_{d,m}$ at updraft velocities of $w \geq 1$ m s^{-1} (Schulze et al., 2020). Our analysis exceeds this former study as we show that *the w threshold above which Aitken mode particles contribute to N_d depends on the properties (e.g., κ, N_{acc}) of the accumulation mode. In addition, we show that various combinations of inferred κ_{acc} and κ_{Ait} result in similar $N_{d,p}$ and thus cannot be constrained without more detailed composition measurements. While these conclusions are drawn on a single observationally-based case study, a more systematic analysis of parameter ranges of Aitken and accumulation mode particles is provided in our follow-up study (Pöhlker et al., 2021).*

Referee 2

This manuscript showed closure results of measured and predicted cloud droplet number concentration for variable updraft speed during ACRIDICON-CHUVA campaign of 2014 where role of updraft speed, hygroscopicity and aerosol size distribution is discussed. Better closure results are obtained when k was assumed to be 0.1, updraft velocity is low and aerosol size distribution is unimodal. CCP and CAS-DPOL are used to measure cloud droplet size distribution and UHSAS with CPC for aerosol size distribution. Updraft speed is measured using Rosemount model 858 AJ probe. Overall the results could be a valuable contribution if they are backed up by proper justification. One of the major point of concern is the lack of reasoning when there is an agreement or disagreement in the closure results. It reads more like a report lacking scientific understanding of the results. I recommend the publication only if the authors improve the discussion part and add previous relevant studies for comparison and show why their approach is better than the earlier studies.

Author response: We thank the referee for the detailed comments and the good suggestions for improving the paper. We have addressed all comments as listed below which significantly improved our manuscript. Referee comments are in black, our responses in blue and manuscript text in *italic* and new text in *red*.

General changes in the manuscript:

1. We clarified how we constrain the measurements on board the HALO aircraft to investigate the relationship between w, N_a, N_d and κ using airborne data and model simulations. This is the first time that such measurements are performed with the proposed methodology ('Probability Matching Method' - PMM). The results from PMM analysis have shown agreement with previous studies and represents a complementary approach in which w, N_a, N_d can be used to constrain CCN hygroscopicity;
2. We revised the abstract and conclusion section to more clearly highlight the new findings and approaches in the current study.
3. More details on the measurements of aerosol and droplet concentrations below and at cloud bases of growing convective cumuli on board the HALO aircraft are are given in terms of time, location and uncertainties (Section 2 and supplemental information);
4. We added statistical parameters to quantify the agreement in the droplet closure.
5. We added a new *Section 4.3: Sensitivities of N_d predictions to w, N_a and κ where we discuss sensitivities of cloud droplet number concentration to κ, N_a and w ($\xi(\kappa)$, $\xi(N_a)$ and $\xi(w)$) in the context of previous sensitivity studies.*

Below you find our specific responses to the referee comments.

Specific Comments:

1 Referee comment: Line 66: Height of the cloud base? Author response: The cloud base heights were different for each region of study. We provide this information for each flight in a new Table in the supplement.

2 Referee comment: Line 63-64: Purpose of using two probes: CCP and CAS-DPOL.

Author response: This was performed to test our methodology with cloud probes that use different characteristics (such as particle inlet, sampling area of detection, sizes sensitivities etc.) to measure cloud particles. We substantially extended Sections 2 and 3 and address the specific referee comment in line 118:

These probes have different measurement characteristics such as particle inlet, sampling area of detection, size sensitivities etc. The CCP-CDP is an open-path instrument that detects forward-scattered laser light from cloud particles as they pass through the CDP detection area (Lance et al., 2010). CAS-DPOL collects forward-scattered light to determine particle size and number that pass the sampling area centered in an inlet shaft that guides the airflow. CCP-CDP and CAS-DPOL has similar values of uncertainty (∼ 10%) in the sample area. However, particle velocities in the sampling tube may be modified by the CAS tube when compared to the open path instruments (like CCP-CDP). This results in an additional uncertainty in the droplet number concentration measured by CAS-DPOL. During the ACRIDICON-CHUVA campaign the resulting uncertainty in the droplet concentration measured by CCP-CDP and CAS-DPOL were ∼ 10% and ∼ 21 %, respectively (Braga et al., 2017a).

3 Referee comment: Line 84 and 186: How is the uncertainty of 30% is estimated?

The uncertainty is actually only 20%, being the sum of 10% uncertainty for aerosol size distribution measurements and 10% uncertainty in the measurement N_d. We noticed that the 30% mentioned in the previous manuscript version were too high as one of the uncertainty was erroneously double-counted.

We added the error bars for $N_{d,m}$ in the figures. To explore the sensitivities of $N_{d,p}$ to an even wider range of N_a, we included now model results for different N_a (±20 %, ±30 %, ±40 %) to not only cover the uncertainty and variability in measurements but also show the sensitivity of N_d to N_a. In addition we expend the discussion about uncertainty and variability of the measured aerosol size distribution. Accordingly, the following changes have been bade in the manuscript (l. 92ff):

The total particle number concentration in the size range of ∼ 10 nm to ∼ 500 nm (N_{CN}) 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). N_{CN} was measured by a butanol-based condensation particle counter (CPCs, modified Grimm CPC 5.410 by Grimm Aerosol Technik, Ainring, Germany) with a flow of 0.6 L min^{-1}. Particle losses in the sampling lines have been estimated and taken into account with the particle loss calculator by von der Weiden et al. (2009). Typical uncertainties of CPC measurements are on the order of ∼10 % (Petzold et al., 2011).

The geometric mean of the aerosol size distribution and N_{CN} below cloud were calculated. The mean aerosol size distribution was fitted by one modal lognormal distributions. The integral of the fit for the aerosol size distribution should be similar to N_{CN} if mainly accumulation mode particles are present. This was fulfilled for AC07, AC09 and AC18, but not for AC19 (Tables S1-S4). For this latter flight, the integrated number concentration of the monomodal lognormal fit made up approximately half of the total N_{CN}. This discrepancy led to the assumption that a significant number concentration of particles in the size range of Aitken mode particles were present during AC19, but not captured by the UHSAS measurements. Consequently, a bimodal ASD shape was inferred. The geometric parameters for the lognormal distribution assumed for measurements during Flight AC19 were based on averages of bimodal aerosol size distributions measured above the ocean in previous studies (Figure S4)

(Wex et al., 2016; Quinn et al., 2017; Gong et al., 2019). The resulting shape of the two modes based on literature data was weighted by the difference between UHSAS and CPC measurements (Table S4). The number concentrations of all fitted aerosol size distributions were normalized to the measured N_{CN}. The variability of the aerosol number size distributions was calculated by the standard deviation on average ∼ 10 % and up to ∼ 20 % for very clean conditions. As a conservative approach ∼ 20 % was used in our model sensitivity study to take into account the impact of this variability on cloud droplet number concentration (Section 4.2). All concentrations are reported for normalized atmospheric conditions (Corrected for standard conditions (STP): T = 273.15°C and p = 1013.25 mbar).

Figure 4 was replaced by Figure R2-1 and the text was adjusted accordingly

4 Referee comment: Line 103: What will be the effect of size dependent hygroscopicity is assumed for external mixing state.

Author response: Assuming external mixing states will result in similar trends of predicted N_d as a function of hygroscopicity. However, previous sensitivity studies have shown that in particular for marine air masses, the assumption of internally mixed aerosol is more appropriate. We added the following text in l. 163ff:

It is assumed that the aerosol particles are internally mixed with identical hygroscopicity (κ) of all particles. This assumption was made based on previous sensitivity studies that have shown that for marine and aged continental air masses internal mixtures are suitable approximations (Ervens et al., 2010).

We also performed additional sensitivity studies in which we assumed different κ values for Aitken and accumulation modes, respectively. The results of these simulations are shown in Figure 3 and discussed in the text (l. 253ff).

To account for different hygroscopicities in Aitken and accumulation modes, we performed further sensitivity analyses using combinations of κ = 0.1 and 0.6 for the two modes (Figure R1-2e). It is obvious that the choice of κ for the Aitken mode (κ_{Ait}) does not affect $N_{d,p}$ for $w \leq\sim 1$ m s^{-1} in the presence of very hygroscopic accumulation mode particles (κ_{acc} = 0.6) or below $w \leq\sim 0.5$ m s^{-1} with κ_{acc} = 0.1, respectively. Even assuming rather extreme values of κ_{Ait} = 0.8 cannot fully reproduce the large increase in N_d at $w \geq\sim 1.5$ m s^{-1} as observed by the CAS probes; assuming very hygroscopic Aitken mode and less hygroscopic accumulation mode particles can approximately reproduce the trend in $N_{d,m}$ from the CDP.

Varying κ_{acc} from 0.1 to 0.6 leads to a large increase of $N_{d,p}$ at all w. The corresponding change in $N_{d,p}$ by increasing κ_{Ait} is much smaller. The reason for this relatively smaller sensitivity of $N_{d,p}$ to κ_{Ait} is the fact that the supersaturation in the cloud is mostly controlled by the droplet growth on accumulation mode particles. The sensitivity of $N_{d,p}$ formed on Aitken mode particles to κ_{acc} is slightly larger if κ_{acc} = 0.1 as compared to κ_{acc} = 0.6, because in the latter case the supersaturation is efficiently suppressed preventing a higher number of Aitken mode particles from activating. Overall we can conclude that assuming different κ values for accumulation and Aitken mode leads to a better representation of the observed trends of $N_{d,m}$ with w (Tables S16 and S17). However, in the absence of more information on the particle hygroscopicity we cannot state with certainty that the assumptions of the two κ values are appropriate for this aerosol population. Figure R1-2d clearly shows that the simplified assumption of a single κ is not appropriate to infer $N_{d,p}$ for low aerosol loading and when the particle number concentrations of the accumulation and Aitken modes are comparable. By using a single κ value, we cannot reproduce the observed continuously strong increase of $N_{d,m}$ for the whole w range. Instead we predict a smaller increase at $w \sim 1$ m s^{-1}, i.e., a flattening of the curve.

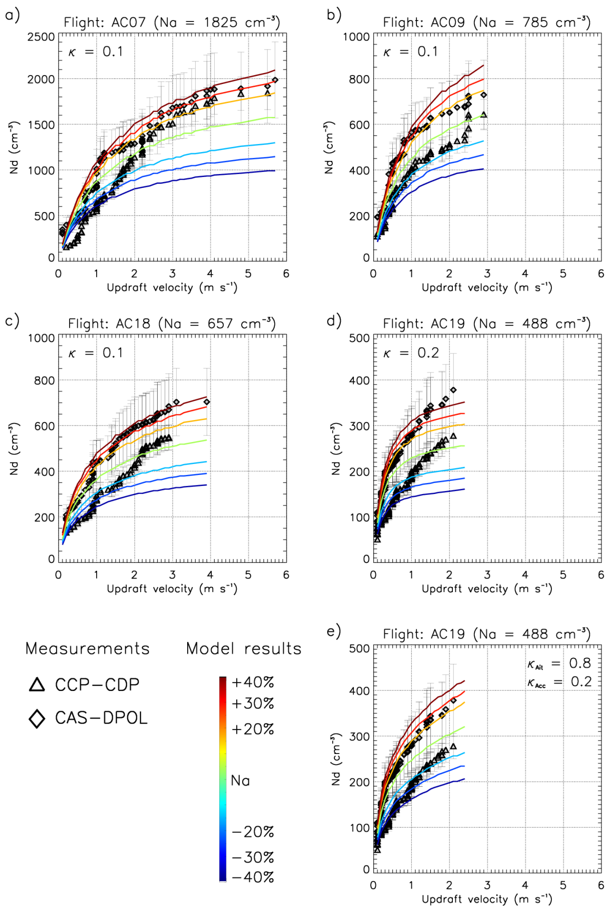

Figure R2-1. Cloud droplet number concentration (N_d) as a function of updraft velocity near cloud base of convective clouds during flights: a) AC07, b) AC09, c) AC18, d) and e) AC19. The measured updraft velocities are based on the "probability matching method" (PMM) using the same percentiles for updraft velocity and $N_{d,m}$ (Section 3.1). The black diamond and triangle symbols represent $N_{d,m}$ near cloud base with the CAS-DPOL and CCP-CDP probes, respectively. Measurement uncertainties (indicated by error bars) are $\sim$ 21% and $\sim$ 10% for CAS-DPOL and CCP-CDP data (Braga et al. (2017a)). The lines show $N_{d,p}$ assuming the uncertainty range of N_a measurements, colored-coded by ΔN_a [%].

5 Referee comment: Line 133: Why there is a deviation between measured and modelled N_d at low w?

Author response: The reason for these deviations is mostly associated with the initial ASD assumed to input the model. For each flight, we have assumed the averaged ASD measured below cloud bases of convective cumuli. This means that it is expected that some disagreement between model and measurements may be found for cloud passes in which N_d were formed from ASDs that the total aerosol number concentration is below or larger than 30% (aerosol concentration uncertainty). Furthermore, cloud passes within pollution plumes from biomass burning may add additional disagreement especially at higher updraft speeds ($w > 2.5$ m s^{-1}).

6 Referee comment: Line 129-130: What is the implication?

Author response: The fact that for all flights a single value of κ can reproduce the measured N_d within all other uncertainties is one of the main findings of our study. We point out that this κ value is an effective value as used in many previous studies to fit the CCN activity. The implications of this findings are that the description of CCN activation and cloud droplet formation for similar air masses can be satisfactorily described by this κ value. However, as we show in our in our additional sensitivity studies (see response to Comment 4), that in the presence of bimodal aerosol size distributions even better closure maybe reached if different κ values for Aitken and accumulation modes are applied we added the following text to the abstract and conclusions:

Abstract: *Above the ocean, fair agreement was obtained assuming an average hygroscopicity of $\kappa \sim 0.2$ (deviations $\leq\sim$ 16%) and further improvement was achieved assuming different hygroscopicities for Aitken and accumulation mode particles (κ_{Ait} = 0.8, κ_{acc} = 0.2; deviations $\leq\sim$ 10%), which may reflect secondary marine sulfate particles. Our results indicate that Aitken mode particles and their hygroscopicity can be important for droplet formation at low pollution levels and high updraft velocities in tropical convective clouds.*

Summary and conclusions: *Above the western Atlantic best N_d closure was achieved for $\kappa \sim 0.2$ applying a single κ value for both Aitken and accumulation modes; an even better representation of the increase in N_d with w was obtained when moderately hygroscopic accumulation mode particles (κ_{acc} = 0.2) and highly hygroscopic Aitken mode particles (κ_{Ait} = 0.8) were assumed.*

6 Referee comment: Line 105: Why collision and coalescence are not considered? Is there any measurement constraint or it is not important to consider?

Author response: Measurement were only performed in non-precipitating clouds. This was checked by CIPgs. We added also more information regarding previous model studies of collision/coalescence (l. 168)

Collision/coalescence processes are not considered as we restrict our analysis to heights near cloud base where droplets are relatively small and the cloud droplet size distribution is narrow. Under such conditions, collision-coalescence is likely negligible (Shaw et al., 1998; Xue et al., 2008; Rosenfeld, 2018; Braga et al., 2017b).

7 Referee comment: Line 147: Under which specific conditions, there will be decrease of N_d due to entrainment of additional aerosols?

Author response: As Referee 1 also questioned the likelihood that entrainment may increase particle concentration and thus cloud droplet concentration, we modified the text in l. 224

~~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)~~. However,while entrainment of biomass burning aerosol may be possible, we do not have any quantitative information on such processes.

We also weakened the corresponding statement in the conclusion section (l. 394):

~~The~~ Our comparison between predicted and measured N_d showed largest discrepancies at high updraft velocities ($w > 2.5$ m s^{-1}), which ~~may be~~ could be possibly explained by non-adiabaticity and/or entrainment of aerosol particles near cloud bases of convective clouds.

8 Referee comment: Line 155: Why is it assumed that bimodal size distribution is due to marine air? Are there any evidences of size dependent chemical composition?

Author response: We agree with the referee that bimodality does not necessarily imply that the air mass had marine origin and vice versa. As it was misleading in the text, we changed it as follows:

The air masses below cloud encountered during flight AC19 were mostly impacted by marine air ~~leading to~~ and exhibited a bi-modal aerosol size distribution with low $N_{d,m}$.

We had strong indications that indeed a monomodal aerosol distribution was not sufficient to explain the observed N_a. We added more details on the fitting of the bimodal size distributions (l. 98ff):

The geometric mean of the aerosol size distribution and N_{CN} below cloud were calculated. The mean aerosol size distribution was fitted by a one modal lognormal distributions. The integral of the fit for the aerosol size distribution should be similar to N_{CN} if mainly accumulation mode particles are present. This was fulfilled for AC07, AC09 and ~~highest during flight AC07 in the southern and northern region of the Amazon Basin (Fig. S1;~~ AC18 but not for AC19 (Tables S1 - S4). ~~Figure 1 shows the measurement region for hte textcolorBrickRedflights analysed in this study~~. For this latter flight, the integrated number concentration of the monomodal lognormal fit made up approximately half of the total N_{CN}. This discrepancy led to the assumption that a significant number concentration of particles in the size range of Aitken mode particles were present during AC19, but not captured by the UHSAS measurements. Consequently, a bimodal ASD shape was inferred. The geometric parameters for the lognormal distribution assumed for measurements during Flight AC19 were based on averages of bimodal aerosol size distributions measured above the ocean in previous studies (Figure S4) (Wex et al., 2016; Quinn et al., 2017; Gong et al., 2019). The resulting shape of the two modes based on literature data was weighted by the difference between UHSAS and CPC measurements (Table S4). The number concentrations of all fitted aerosol size distributions were normalized to the measured N_{CN}.

9 Referee comment: Line 182-183: The sensitivity analysis of Aitken and accumulation mode to total N_a and N_d should be included in this study as this is one of the highlight of this manuscript that represents scientific advancement.

Author response: We agree with the referee that our data set showed interesting results that highlight the possible importance of the individual properties of Aitken and accumulation mode particles for cloud droplet number concentration.

In the revised manuscript, we added some findings from our recent study (Pöhlker et al., 2021) that was published in ACPD nearly concurrently with the present article, and was just accepted. There we show for example, that the sensitivities of κ and N_a to N_d are different for accumulation and Aitken mode particles, respectively. This was added in line 278:

In our recent model study, we have shown that in the transitional regime, i.e., in the parameter space between the aerosol- and updraft limited regimes, as defined by Reutter et al. (2009), N_d can be equally sensitive to κ and w (Pöhlker et al., 2021). In that study, we show that with increasing N_a, the sensitivities to both parameters decrease; however, the sensitivity of N_d to w remains higher under such conditions than that to κ.

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