Author response to referee comments

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 **??**e). 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 **??**d clearly shows that the simplified as-*

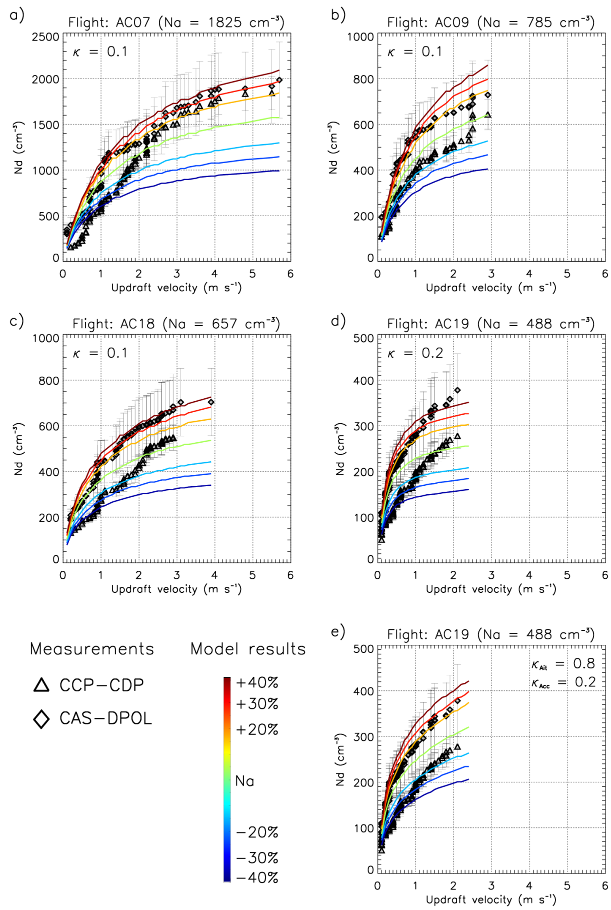

Figure R2-1. Cloud droplet number concentration (N_d) as a function of updraft velocity near cloud base of convective clouds during flights: a) AC07, b) AC09, c) AC18, d) and e) AC19. The measured updraft velocities are based on the "probability matching method" (PMM) using the same percentiles for updraft 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 [%].

sumption 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.

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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