Author response to Referee 3's comments

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*.

Below you find our specific responses to the referee comments.

Referee 3

The main value of this work lies in the information offered from measurements of aerosol and cloud properties in fairly convective clouds in an interesting location.

1) The main objective(s) of this paper are unclear: a. The title indicates closure, but reasonable closure is not possible here for two reasons. One is that there are two measurements of cloud droplet number concentrations that are separated by 20-30% before considering the uncertainty in the measurements. The second reason is that there is no independent estimate of kappa. With the information presented, the model is incapable of producing closure because it is the comparison of the model and observations that is used to estimate the best kappa. This implicitly assumes that the other observations are correct, but we know that is not the case because the two Nd measurements disagree. I am not being critical of the Nd measurements: I applaud the authors for including both. My suggestion is to take your best case, in terms of measured quantities, including information available to define kappa, and attempt closure of the model with the two measurements of Nd. Your best case might be the simplest, and the one that has the best estimate of updraft speed or w; more discussion of w later. The objective of this best case would be to be able to say something about how the two measurements relate to the modelled Nd.

Author response:

(a) The focus of the paper is to describe the comparison of measured and modeled droplet concentrations at cloud bases of convective clouds. We describe a new approach using a different set of in situ measurements as it has been done before and a parcel model.

(b) We respectfully disagree that the term 'closure' is not appropriate here. We refer to the definition of 'closure', as described in previous studies, e.g. by Quinn and Coffman (1998): *"In a closure experiment, an aerosol property is measured by one or more methods and calculated from a model that is based on other independently measured properties. A comparison of the measured and calculated values can reveal inadequacies in either the measurements or the model."* Even the aforementioned study did not focus on cloud droplet number concentration, generally this definition is very well in agreement with our approach.

(c) The closure analysis was performed separately for each cloud probe in order to verify the methodology using two types of instruments. The biases of the results from both probes were expected since they have differences in measurements characteristics and uncertainties. Furthermore, as shown at Braga et al. (2017) a perfect agreement is not expected between them since they were mounted in different wings and measured different air samples. Braga et al. (2017) have shown that both probes agrees within their measurement uncertainties. This is an inherent characteristic of in situ measurements. Our study reveals the sensitivity of N_d to various parameters (κ, N_a, w). To our knowledge, there are not many studies in the literature that performed comparable studies for a such a rich data set and therefore could be used to reveal sensitivities. The statistical results that are well described for both probes within the main text and supplement.

(d) We realized that some of your text was misleading as we do not fit our results to κ to get a 'best estimate' but rather describe our results with the adiabatic parcel model as a function of prescribed κ values that are in the range of previously determined ones. The novelty of our analysis is the possibility to assess the relative uncertainties of various parameters based on a unique combination of measurements.

We make these points clearer throughout the revised manuscript.

b. In the abstract and later in the text, there is considerable discussion of the best kappa value. I don't see this as a major objective of this paper. There are better ways to estimate kappa, which the authors refer to, than to use such a convoluted approach that includes uncertainties. Use estimates of kappa from previous studies in the region.

The focus of the paper is to describe the closure analysis at cloud bases of convective clouds. The referee is right that indeed there are more straightforward ways to estimate κ but most of them do not use data from real cloud bases for such a broad range of conditions as we did. We do not suggest methodology is better to constrain κ and we agree with the referee that our method is even more complicated and 'convoluted' than others. However, this is in fact one of the main points of our paper to demonstrate the uncertainties in measurements and how they translate into predictions of cloud droplet number concentrations. To our knowledge there are not many studies available that use such a rich set of measurements (e.g. two sets of N_d, w analysis) and perform a detailed analysis of their importance for N_d prediction at cloud bases.

c. The last sentence of the abstract says that "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." This seems like a result worthy of publication, if the authors first acknowledge previous related work: Leaitch, W. R. et al., Effects of 20–100 nm particles on liquid clouds in the clean summertime Arctic, Atmos. Chem. Phys. 16, 11107–11124 (2016); Baccarini et al., Frequent new particle formation over the high Arctic pack ice by enhanced iodine emissions, Nature Comm. https://doi.org/10.1038/s41467-020-18551-0, 2020.

We thank the referee for these additional references. We would like to refer to our recent publication that includes a detailed discussion on conditions under which Aitken mode particles contribute to CCN (Pöhlker et al., 2021). While the referee is right that under Arctic conditions, i.e. when N_a, this might be the case, the parameter space is much broader as it depends on combination of κ, N_a and w. Since the conditions in the Arctic are substantially different to those in our current study in terms of aerosol loading, w and CCN-limited regime, we did not add the references to the manuscript.

d. The last sentence of Section 1 tells us that the modelling is used to examine the sensitivities of Nd to kappa, Na and w, but we've been subjected to such sensitivity studies for many years now, dating back to at least 1971 (Junge and McLaren, JAS, 1971), which told us that the number distribution was more important than the chemical content of the particles. Why is another sensitivity study of this important?

The referee is right that the relative importance of aerosol parameters for cloud droplet number concentration as identified in this study is not new, as it is discussed in Section 4.3. Many of the studies cited in this section are based on idealized cases or conditions.

The novelty of our study is the application of the unique combination of measurements. While at a first glance the two N_d measurements disagree substantially, we demonstrate that both measurements can be reconciled by using a new method that take the advantage of new in situ measurements below and within convective cloud bases. Such type of analysis were not performed as it is presented in this manuscript and over the Amazon basin.

2) Lines 42-50: Leaitch et al., Cloud albedo increase from carbonaceous aerosol, Atmos. Chem. Phys., 10, 7669-7684, 2010 is relevant here.

We thank the referee for this additional reference and added it in the new version.

3) Line 93 – What is the Aerosol Measurement System, and why do we need to know its acronym?

This acronym has been used regularly for data from the HALO aircraft. Thus, we also use it here to make our data sources as transparent as possible and so that the reader can make the connection to other data from the HALO database. We reworded the paragraph as follows to make it clearer that this measurements system measured the total aerosol particle number concentration as used in this study (l. 100):

The total particle number concentration in the size range of ∼ 10 nm to ∼ 500 nm (N_{CN}) below cloud base was measured using the Aerosol Measurement System (AMETYST). The system is composed of scanning mobility particle sizers and butanol-based condensation particle counters (CPCs, modified Grimm CPC 5.410 by Grimm Aerosol Technik, Ainring, Germany) with a flow rate 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; Andreae et al., 2018).

4) Lines 104-106 – Your inferred Aitken distribution has a minimum at about 80 nm, based on the Wex, Gong and Quinn distributions. The below-cloud distributions measured over the North Atlantic Ocean by Leaitch et al. (ACP, 2010), also used in parcel model calculations, had minima at about 100 nm. Such a minimum might make a substantial difference in your results, for example, potentially requiring a smaller kappa for the Aitken mode.

Many thanks for this comment. In our recent publication by Pöhlker et al. (2021) we indeed showed a large sensitivity of the mode diameter of the Aitken mode to the activation of Aitken mode particles. However, we are confident that the minimum between the two modes in the current study was below 92 nm; a minimum at larger sizes would have shown a different shape in the UHSAS measurements. We added the following text (l. 114):

Other shapes of marine aerosol size distributions, e.g. as reported by Leaitch et al. (2010), were not considered for our lognormal fit because they were not in agreement with the measured UHSAS data.

5) Lines 118-126 – Are the differences in the sampling approach by the CAS and CCP potentially responsible for the bias in the two measurements?

Yes, but this is not the only reason. As mentioned above the probes were also mounted on different wings, ∼20 m apart from each other. We performed a detailed analysis about the measurements of CAS and CDP in our previous study Braga et al. (2017). Our results indicated that the agreement between CAS and CDP is good assuming the probe uncertainty ranges.

6) Lines 131-133 – Given the better agreement with the King probe, why were the CAS measurements not chosen to compare to the model alone? Were number or sizing differences mostly responsible for the differences between the CAS and CCP probe comparisons with the King probe?

We are somewhat confused about this suggestion as it apparently contradicts the referee's introductory comments (*I am not being critical of the Nd measurements: I applaud the authors for including both.*. We do agree with this referee comment that the consideration of both data sets our study apart from others that base their conclusions from N_d closures on only one date set.

The CAS measurements had better agreement with the King probe due to the fact that the hot-wire was mounted at the CAS probe, and thus, measured the same air sample. Furthermore, the CDP is an open-path instrument, while CAS measure drops that pass the sampling area within a pitot tube. Such characteristics lead to differences in particle sampling, size sensitivities etc. Nevertheless, both probes measured realistic values and the fact that the closure analysis have shown similar results for most of flights enhances the relevance of the use of two probes.

Usually, in other studies, measurements of only one of the probes are available. Thus, our study shows (i) that N_d measurements from different probes might result in different values and (ii) these differences do not translate into significantly different conclusions on the relative importance and absolute values of aerosol parameters to reach N_d closure.

7) Section 3.1 – a) It would be helpful to have an example of the time series of Nd and w during a cloud penetration to demonstrate the application of this approach here. By design, the PMM analysis appears to ensure the connection between Nd and w shown in Figures 3 and 4, whereas other methods, such as Peng that you later refer to as weaker (Lines 339-345), do not. Because of the apparent forced dependence by the PMM method, it seems inappropriate to use this approach to make conclusive statements about the dependence of Nd on w.

We have added a new figure showing the time series of N_d and w within cloud. The new text and figure are shown as below (l. 166):

"...Figure 2 shows an example of measured w at cloud bases and estimated w based on the PMM analysis (w_{PMM}). The figure shows that w_{PMM} are in well agreement with measurements at cloud bases. Furthermore, for cloud passes in which the values of w are negative realistic w are estimated based on PMM."

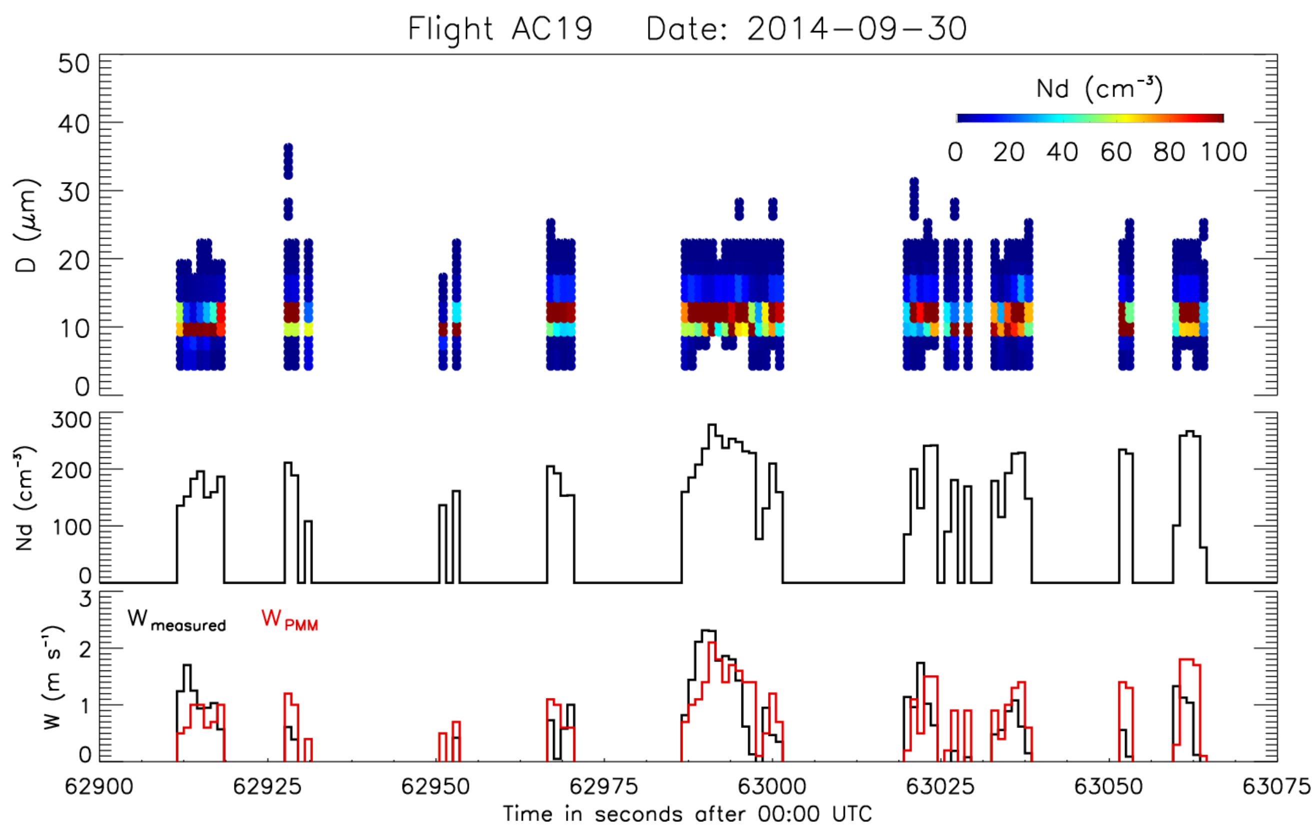

Figure R2-1. Time series of droplet size distribution measured by CCP-CDP [top], Number concentration of droplets (N_d) [middle], Measured vertical velocities w, and estimated w based on PMM method [bottom]. The measurements were performed during flight AC19 above the Atlantic Ocean.

b) The Peng approach, and perhaps others, were developed for stratus and stratocumulus with relatively low updraft speeds. The pathways of air parcels through such clouds, as shown by modelling studies (e.g., at CSU), are much different than for more convective clouds, and therefore the Peng approach may not be appropriate here in any case. For strong convection, the w may significantly increase with height above cloud base. How do you know that your w, measured more than 20 metres above base, were indeed representative of the w at cloud base? The Nd may not change much with height (assuming only homogeneous entrainment), but the w can, and based on the LWC shown in Figure 2 sampling was conducted well above cloud base in many cases, which would lead to overestimating w in some cases.

For cloud base height we assumed the level of maximum supersaturation from the vertical profile of simulated supersaturation. Since the value of supersaturation over water within cloud cannot be measured, the height above cloud base mentioned in the MS regards to model simulation results.

The measurements of w and Nd were performed at cloud bases of cumulus humilis and mediocris. These cloud passes were selected based on the videos recorded by the HALO cockpit forward-looking camera. Therefore, we could not ascribe a specific height for measurements of cloud bases but rather state that they had approximately the same altitude during the cloud passes.

Figure 2 shows the simulated and measured values of LWC. The heights of LWC are only shown for the model results. The frequency of measured LWC is shown for cloud bases regardless of the height above cloud bases.

We clarify these points at Section 2.2 as follows:

"*...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). The cloud base measurements were selected based on the videos recorded by the HALO cockpit forward-looking camera...*"

We have re-written section 3.2.2 to better explain this point (l. 187 ff):

The cloud base measurements were performed at approximately constant altitude during each research flight and were selected based on the videos recorded by the HALO cockpit forward-looking camera. However, these measurements might represent different levels in relation to the level of maximum supersaturation at cloud base, which depends on the updraft velocity and turbulence in cloud. In order to determine the height at which $N_{d,m}$ and $N_{d,p}$ should be compared, the measured liquid water content (LWC) was compared to the simulated LWC using the aerosol size distribution for the different flights, together with w measured at cloud base and an assumed hygroscopicity of κ = 0.1.

Under adiabatic conditions, $N_{d,p}$ is approximately constant at $\sim$ 20 m above the level of the maximum supersaturation S_{max} (Fig. S5). Figure 2 shows the predicted LWC from the simulations as a function of height above S_{max} for the four flights. Overlaid on the model results (colored lines) are the frequencies of measured LWC by the cloud probes near cloud base (white bars). The measured LWC represents the cumulative mass size distribution. For all flights the model predictions in most of cases match the minimum LWC measured at $\sim$ 20 m above the S_{max} level. This height level might represent slightly different absolute heights above the surface and the level of saturation estimated by the model (RH = 100%) (Fig. S6). However, since we focus our discussion in the following section on the comparison of $N_{d,m}$ and $N_{d,p}$, we perform our analysis assuming modeled data for a height of 20 m above S_{max}.

8) Line 190 – Unless there is some inhomogeneous mixing.

The referee is correct that inhomogeneous mixing in cloud might lead to a reduction in N_d. However, in the text here,we refer to our model predictions. Inhomogenous mixing cannot be taken into account in our adiabatic parcel model.

9) Lines 207-208 - Curious statement: effectively, you are saying that variations in Nd of 20-40% aren't large enough to worry about. Is that the status of the indirect effects?

We do not fully understand the referee's comment. We stated in the text that the two sets of N_d measurements differ by $\sim$20% and that these values can be reproduced by the model by using very similar κ values. In our opinion, this is fully in agreement with the initial referee comment and our response (1.d) and with many previous studies that are cited in Section 4.3 that have shown a weak sensitivity of N_d to κ.

Since we feel that sensitivities and this parameter ranking are sufficiently discussed in the introduction and in Section 4.3, we did not change any text here.

10) Lines 209-211 - This statement is predicated on the correct answer lying between the two measurements, yet there is nothing in the paper to suggest that is correct.

We did not mean to imply that the 'best value' is in-between those of the two measurements. In fact, we had used averaged values in the previous version of the manuscript (but were rightfully criticised by the referees for doing so). Therefore, we performed our closure study for both measurements separately. As we realized that the text might be misinterpreted in this regard, we changed it as follows (l. 221):

The use of a single cloud probe might lead to a biased estimate based on the data set of each cloud probe separately. The consideration of both cloud probes shows the uncertainty in N_d measurements and therefore the uncertainty range of κ and/or N_a values for N_d closure.

11) Lines 219-220 – "Confirm" is an exaggeration. It could be true, but you haven't demonstrated this, and there are better ways to estimate kappa. This approach is too uncertain.

We agree with the referee that our approach of deriving κ may be more uncertain than others, such as CCN (as opposed to N_d) closure studies or direct composition measurements. However, given that we derive similar conclusions as such previous studies suggests that our measurements of w and N_a and their correlations as implied by the PMM approach were sufficiently constrained. We reworded the text as follows (l. 229): *Our results ~~confirm~~ are in agreement with previous studies. The value range is representative of internally mixed aerosol particle populations during the dry season in the Amazon Basin, which are influenced by fresh and aged biomass burning aerosol from Amazon and Africa.*

12) Lines 222-225 – If you sampled very near cloud base, then it would seem difficult for entrainment to increase the Nd. Again, it is important to illustrate your in-cloud selection process. If you sampled too high above cloud base, then you need to question your estimate of w. Also, your statement here is incorrect if the CCP measurements of Nd are closest to reality. Overall, I don't find this paragraph useful.

We agree with the referee that this text distracted from our main messages. Therefore we removed it.

13) Lines 231-235 – When you say the hygroscopicity of particles could change due to dissolution of soluble compounds, do you mean the uptake of highly soluble gases, such as HNO3? If so, a reference is in order: Kulmala et al., maybe JGR, in the 1990s. If you are talking about delays associated with weakly soluble compounds, then Shantz et al.: Effect of organics of low solubility on the growth rate of cloud droplets, J. Geophys. Res., 108, 4168-4177, 2003 is appropriate here and on line 405. Your assumption that reduced solubility might flatten the curves more may or may not be correct, depending on the entire chemical composition of the size distribution. Shantz et al found that delayed growth reduced the Nd relative to a highly soluble compound. I don't see enough support for your statement that "such composition effects can likely be excluded."

We agree with the referees that there are several aspects that would need to be considered for any speculations regarding such composition effects. Therefore, we removed these two sentences and rather frame our conclusions in the context of an 'effective κ' as it has been done in previous studies and that encompasses several composition effects.

14) Lines 236-237 – Yet, your Aitken mode is highly soluble.

We do not have any information on the solubility of the Aitken mode particles. However, we do show that various combination of κ_{Ait} and κ_{acc} can lead to similar $N_{d,p}$. The sentence as written does not exclude the possibility that Aitken mode particles are equally or more hygroscopic than accumulation mode particles.

15) Lines 338-247 – Again, I suggest that your tendency of measured Nd vs w is at least in part a result of your approach.

Yes, our approach, i.e. the PMM, implies that there is a correlation between N_d and w. We did not make any changes in the manuscript as the method is explained in Section 3.1 and in more detail in previous publications (Haddad and Rosenfeld, 1997; Braga et al., 2017a)

16) Line 274 – Nd to Na?

Thank you. The referee is right that the text should read:

...it was demonstrated that the sensitivity of N_d to N_a becomes small...

17) Lines 350-353 – "we conclude that the sensitivity of Nd to Na is much greater than that to w under these conditions which is also reflected by the rather small increase in Nd with w at high updraft velocities". Again, we need to know where with respect to cloud base that the w were measured, in case some of your w are overestimated.

We think the referee misunderstood where the measurements of w and Nd took place. We tried to better explain that the measurements of w were performed within cloud bases selected based on the videos recorded by the HALO cockpit forward-looking camera (as described in the new Section 3.2.2 and in our response to Comment 7). The exact height of measurements above cloud base (Smax) cannot be estimated. The height above cloud base (S_{max} level) of 20 m was used for modeled N_d for the reasons described in Section 3.2.2. This section was modified now to better explain our methodology to constrain modeled and measured N_d at cloud bases.

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