Author response acp-2021-80

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Author response: We thank the referee for the detailed comments. Below you find our specific responses (in red) to the referee comments (in black). Manuscript text is in purple, new text in purple bold.

Anonymous Referee #3

1- Authors' response to my original comment 1): "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."

Review comment on this response: - Of course, I agree with the stated reference as to the nature of closure. Unfortunately, I find this statement to reinforce my point, rather than the authors. You have not measured kappa by any methods. You use the model to make an assessment of kappa, but you cannot call it closure because you do not have anything to give you an independent assessment of kappa to compare against.

Author response: The reviewer is correct that we did not use direct kappa measurements that were performed during the same field campaign. However, as we point out throughout our manuscript, we used kappa values from other studies at the same location and in other comparable air masses (e.g. marine). These kappa values were determined independently from the measurements of the other aerosol properties, namely particle number concentration.

Our model was NOT set up such that we fitted kappa to get the best agreement of measured and modeled cloud droplet number concentration without any constraints on the kappa value. In that case, we would agree with the referee that such a model exercise should not be called 'closure', but 'finding the best fitted value'.

The assumptions made in our model studies were guided therefore by previous measurements as indicated throughout the manuscript, e.g.

l. 11: assuming an average hygroscopicity of $\kappa \sim 0.1$, which is consistent with Amazonian biomass burning and secondary organic aerosol

l. 227 ff: This κ value has been suggested previously for comparable air masses during the dry season in the Amazon Basin (e.g., Pöhlker et al., 2016, 2018). In these prior studies, κ was constrained based on size-resolved CCN measurements and measurements of the aerosol chemical composition, dominated by an aged organic fraction. [...] 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.

l. 270: in the absence of more information on the particle hygroscopicity we cannot state with certainty that the assumptions of the two values are appropriate for this aerosol population.

2- Authors' response to my original comment 1 b): "The referee is right that indeed there are more straightforward ways to estimate kappa but most of them do not use data from real cloud bases for such a broad range of conditions as we did."

Review comment on this response: Why would there be a difference between estimating kappa below a cloud rather than not below a cloud? Effects on photochemistry might play some role, but I don't see how your results that show a range of kappa values is either better or for that matter distinguishable from an approach that offers a more direct estimate of kappa, whether below or not below a cloud.

Author response: We are afraid that the referee misunderstood our previous response and the single sentence he cites was taken out of the context. We do not imply that the kappa values below and in cloud are different. (Even though this may be the case due to dissolution of soluble particle components or other effects but this is not topic of our study.)

For completeness, we repeat our complete response to the referee's previous comment here.

"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 Nd, w analysis) and perform a detailed analysis of their importance for Nd prediction at cloud bases

Based on this, we would like to re-emphasize the novelty of our study to use **cloud droplet measurements, paired with fairly well constrained updraft measurements.** Most of the past closure studies were performed using CCN measurements in CCN counters, i.e. at equilibrium conditions. Such measurements mimic conditions below cloud, i.e. when particles take up water vapor but have not grown to cloud droplets yet.

Compared to such *CCN closure studies*, the number of *cloud droplet number (Nd) closure studies* is much smaller since cloud properties, such as drop number concentration and co-located updraft velocities, are much more challenging to measure. However, by our analysis, using PMM, we reduced the uncertainty in such Nd closure studies by better constraining w and therefore reducing the uncertainty.

This is stated, e.g. in l. 409ff: *Implying that higher Nd are formed in regions of higher updraft velocities, we sorted observed data of Nd and w by their frequency of occurrence ('probability matching method'). Using this approach, we reduced the uncertainty of w for the Nd closure. Therefore, we could largely limit our sensitivity analysis to the investigation of the importance of particle hygroscopicity and number concentration for cloud droplet number concentrations.*

3- Authors' response to my original comment labelled 1c: "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."

Review comment on this response: This is about processes, not location. The relative importance of the many relevant processes may vary from one location to another, but in this case one of the most important factors is the effect of "aerosol loading". Low concentrations of larger particles are the main reason Aitken particles can activate in the Arctic. As for updrafts, if particles as small as 20-30 nm can activate in the Arctic, then it is more likely to happen in cases of higher updrafts. These references support your work here, and they are warranted.

Author response: The referee is correct that our response was not really clear. Of course, the conclusions regarding a possible contribution of Aitken mode particles to CCN is not depend on location but on the parameters and processes.

In our previous paper (Pöhlker et al., 2021), we discuss in detail that the role of the Aitken mode particles depend on a combination of aerosol loading and updraft velocity. We would like to point out that our study focuses on cumulus clouds whereas the studies the referee cites here are on stratus clouds for which the Na/w thresholds are different above which Aitken mode particles contribute to CCN.

To make this clear, we added in l. 366 (new text in bold)

Qualitatively this was also suggested in a previous Nd closure study for marine stratocumulus clouds, where it was concluded that only the presence of an Aitken mode could explain the high Nd,m at updraft ***velocities of w ≥ 1 m s-1 (Schulze et al., 2020)). Generally, the conditions at which Aitken mode particles contribute to CCN depend on the combinations of the parameter values of Na, w and*** ***κ (Pöhlker et al., 2021). Therefore, Aitken mode particles were shown to contribute to CCN in Arctic stratocumulus clouds or fog, that are characterized by low w and (Jung et al., 2018; Korhonen et al., 2008; Leaitch et al., 2016) whereas both updraft and aerosol loading are much higher in the convective cumulus clouds in the Amazon.***

4- In response to review comment 7b), the authors state: "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".

Review comment on this response: Thank you for Figure R2-1. You should be able to calculate an approximate distance above cloud base based on the size of the droplets using your adiabatic parcel model. LWC is dependent on height above base, not updraft speed, so you just need to define the activated number concentrations and see at what heights you reach your measured diameters. You don't even need a lot of sophistication, and this will offer a better estimate of height above base.

Author response: We thank the referee for their suggestion. This is exactly the procedure we applied in order to constrain the height in cloud at which we compared the measured and predicted cloud droplet number concentration:

- Using our parcel model, we calculated the LWC based on the predicted droplet number concentration and droplet size.

- The resulting LWC was then compared to the measured LWC

- The predicted height above cloud base using our adiabatic parcel model at which both the predicted and measured LWC showed best agreement was then defined as the 'approximate distance above cloud base' at which our closure was performed.

We did not perform a comparison of predicted and measured cloud droplet sizes but, other than that, it seems that our approach is what the referee was suggesting and which led us to the conclusion that a height of ~20 m above cloud base is an appropriate value. Since we dedicated a full Section (3.2.2 Determination of in-cloud height to compare Nd,m and Nd,p) and Figure 3 to this, we did not add any further description to the text.

5- In response to review comment 14 ("Yet, your Aitken mode is highly soluble"), the authors state that they have no information on the solubility of Aitken mode particles. On lines 259-261 of the revised paper, they state that "Even assuming rather extreme values of kappaAit = 0.8 cannot fully reproduce the large increase in Nd at w & 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 Nd,m from the CDP."

Author response: We are not fully sure that we understand the referee's comment. Does the referee would like to point out an apparent contradiction? We do not conclude that the Aitken mode was indeed highly soluble. It should be kept in mind that we use kappa as an "effective parameter, encompassing all factors that affect water uptake" (l. 177, and in agreement with previous studies). To make this clearer, we added at l. 261 (new text in bold)

Even assuming rather extreme values of Ait = 0.8 cannot fully reproduce the large increase in Nd at w ≥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 Nd,m from the CDP. ***It should be kept in mind that κ is considered an effective parameter that may also reflect water uptake due to additional processes or effects that are not represented in our model and therefore cannot be further reconciled here.***

6- Review comment 15 – The authors acknowledge that the approach they use to associate Nd and updraft speed reinforces a strong correlation. In other words, the approach is not entirely objective. This should at least be discussed somewhere in the paper.

Author response: The referee is correct that the Probability matching method (PMM) applied here implies a correlation of Nd and w. Strictly, this approach is therefore indeed not entirely objective but implies the assumption that one of the parameters affects the other. We do not think that we need to defend this assumption as it follows from the equation that is generally accepted as a basis for cloud physics (Twomey, 1959).

$N_{CCN} = N_0 \cdot S^k$

whereas the supersaturation S generally increases with updraft velocity (with all else being equal). Deviations from this relationship may be caused, for example, due to entrainment. However, near cloud base this is unlikely. While this had been discussed in the cited references (Braga et al., 2017), it was not clearly pointed out in the current study. Therefore, we added in l. 160 (new text in bold):

The PMM analysis is based on the assumption that these two related variables increase monotonically with each other. ***This assumption implies that entrainment – which may lead to a reversal of the assumed trend - can be neglected near cloud base which is likely a valid assumption under these conditions.***

7– I do applaud including both measurements of Nd, but I feel that one is likely to be closer to the truth than the other, and I think this could be assessed.

Author response: We are not sure what the referee is proposing here. Based on our analysis, we cannot assess whether one Nd measurement is 'closer to the truth' than the other. In fact, we are not even sure how to define 'truth' in this context since the two probes use different characteristics to determine the droplet number (cf. Section 2.2). However, we are convinced that reporting data from both probes is very valuable as it demonstrates that there may be biases in conclusions if only data from one of the probes are considered. Since this was also emphasized by the referee in their last report and repeated here ("*I do applaud including both measurements of Nd") and also made clear throughout paper, e.g.*

l. 63: This [i.e. the closure analysis] was performed to verify our methodology using two types of instruments to measure number concentrations of droplets with different particle inlet characteristics and uncertainties

l. 219: The deviations between Nd,m from CCP-CDP and CAS-DPOL (~ 21% on average) reinforce the advantage of duplicate measurements for the closure analysis. The use of a single cloud probe might lead to a biased κ estimate based on the data set of each cloud probe separately.

References

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