

**Responses from authors to reviewers of the manuscript:** “Further evidence for CCN aerosol concentrations determining the height of warm rain and ice initiation in convective clouds over the Amazon basin.”

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### **General comments**

We appreciate and thank the Editor for his considerable effort and care in the complex review of this paper.

The authors thank the referees for the general comments and advices. Furthermore, the advices of the referees are highly appreciated as well as the very valuable and constructive suggestions to increase the quality of the manuscript. We tried to address the points requested by the reviewers to the paper be considered for publication. Overall, we have improved the focus of the paper highlighting our objectives and the novelty of our study.

In the new version of the manuscript we have removed two Figures (3-4) as requested by the reviewers and an additional schematic Figure which summarizes our findings was added in the discussion section.

In the following text the reviewers' comments are highlighted in blue color and the authors responses are provided below.

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## Referee number 1

### Report 1:

The authors have not satisfactorily addressed several of the very reasonable and serious points made by the referees and editor. As far as I can tell, the revised manuscript has changed very little. Many of the problems listed in the first review remain in this manuscript.

These include:

1. The main result of the paper is to produce a relationship between the height of the first measurement of drizzle drops and the estimated number of cloud drops at cloud base (Fig 17). The problem with the approach and the resulting relationship is the lack of attention to detail. The authors have offered very little discussion of the limitations of the observations and processes which can break or increase the error in the simple relationship. Ultra-giant aerosols, enhancement of collision and coalescence due to turbulence and increased supersaturation due to entrainment and mixing will all potentially weaken the relationship.

A: All these factors were not measurable. In the case of giant CCN we found no evidence for large concentrations (as shown at Figure 10), but the observation of rain initiation for  $r_e \sim 12 \mu\text{m}$  (see Figure 9a) is an indication of the presence of GCCN (this was also observed at previous studies e.g. Freud and Rosenfeld, 2012 and Konwar et al., 2012).

The observed results mean that all the processes that are the subject of the reviewer's concern are secondary in their importance to the process that determines the documented relationships, as described in the manuscript. We add the text below in the manuscript.

“The linear relationship between  $N_a$  and  $D_{13}$  indicates a regression slope of about  $5 \text{ m (cm}^{-3}\text{)}^{-1}$  between  $D_{13}$  and the calculated  $N_a$  for the Amazon during the dry-to-wet season. This value is slightly larger than the values observed by

Freud and Rosenfeld (2012) for other locations around the globe (e.g., India and Israel). These clear linear relationships found between  $N_a$  and  $D_r$  ( $\sim D_{13}$ ) for different regions highlight the efficiency of the adiabatic parcel model to estimate the height of rain initiation within convective clouds. Additional cloud processes associated such as GCCN, cloud and mixing with ambient air and other processes that are not accounted for in this study produce deviations which are already included within the observed uncertainty of the linear relationship  $N_a$ - $D_r$ .”

These very concerns of the reviewer render the scientific significance the findings of this paper as quite high, because the reviewer was concerned that these other processes would mask the relationships between  $N_a$  and  $D_r$ , but in fact they do not. This realization worth publishing.

R1: 2. There are no figures in the paper to show that the measurements were made in the main updraft near cloud top to avoid observing raindrops formed at higher levels and falling in the downdrafts around the cloud edges. What is the evidence that the cloud top was not higher at an earlier time? In fact there are no figures showing the dynamical structure of the clouds.

A: Figure 2 illustrates the flight pattern near cloud tops. It is documented in the videos of the nose camera. Again, if there was a risk of rain falling from above, which is minimized as we described in the manuscript, it would decreased the strength of observed relationships. To the extent that the data was contaminated by rain from above, it only demonstrates that this did not happen to the extent of masking the relationships, which is independently hypothesized and observed previously, as described in section 3.1. See below the relevant texts from the manuscript.

“...It is assumed that rain (or ice) formation starts when calculated DWC exceeds  $0.01 \text{ g m}^{-3}$  (Freud and Rosenfeld, 2012). For rain initiation in liquid phase the DWC threshold is  $\sim 10\%$  greater due to the overestimation of DWC during CIP measurements in warm clouds (as stated at Section 2.2.1). The small terminal fall speed of the drizzle drops ( $\leq 1 \text{ m s}^{-1}$ ) allows to focus on in-situ rain (or ice) initiation while minimizing the amount of DSDs affected by rain drops fallen from above into the region of measurements. In addition, cloud passes with rain were eliminated when cloud tops were visibly much higher than the penetration level ( $> \sim 1000 \text{ m}$ ), based on the videos recorded by the HALO’s cockpit forward-looking camera. However, cloud tops higher than few hundred meters above the penetration level occurred only rarely.”

R1: 3. There is the persistent argument that the almost constant value of the effective radius is evidence of inhomogeneous mixing...

A: The reviewer misses the point that inhomogeneous mixing leads to adiabatic effective radius, and not a fixed one. The fact that the measured  $r_e$  is close to the estimated adiabatic  $r_e$  highlights the observations that the behavior of cloud mixing with air is nearly inhomogeneous, and therefore the effective radius behaves nearly as in adiabatic cloud. This is explained better now at section 3.2.

R1: If inhomogeneous mixing had occurred, there would likely be an increase in supersaturation and hence in the size of the largest drops as well as activation of CCN from the drops evaporated due to the mixing. Importantly, the raindrops would most likely form at a lower level than in a cloud region containing the number of cloud drops activated at cloud base and moved in an adiabatic parcel. Similar arguments can be made for turbulent enhancement.

A: The main process that we highlight as responsible for the relationships between  $N_a$  and height for rain initiation ( $D_r$ ) is evidently the dominant one. Additional processes such as turbulence etc. produce deviation which are already included in the uncertainty of  $N_a$  and  $D_r$  relationship. We add the text below to the manuscript.

“The linear relationship between  $N_a$  and  $D_{13}$  indicates a regression slope of about  $5 \text{ m (cm}^{-3}\text{)}^{-1}$  between  $D_{13}$  and the calculated  $N_a$  for the Amazon during the dry-to-wet season. This value is slightly larger than the values observed by Freud and Rosenfeld (2012) for other locations around the globe (e.g., India and Israel). These clear linear relationships found between  $N_a$  and  $D_r$  ( $\sim D_{13}$ ) for different regions highlight the efficiency of the adiabatic parcel model to estimate the height of rain initiation within convective clouds. Additional cloud processes associated such as GCCN, cloud and mixing with ambient air and other processes that are not accounted for in this study produce deviations which are already included within the observed uncertainty of the linear relationship  $N_a$ - $D_r$ .”

R1: 4. As the authors admit themselves, they have insufficient information to examine ice initiation in these clouds.

A: We show where first ice is found in CIP images. The initiation of ice can be visually ascribed for sizes greater than  $\sim 0.25$  mm and it does not mean that frozen smaller particles cannot be present. This is commented at the manuscript as shown below:

“Table 3 shows the cloud depth above cloud base at which warm rain initiation ( $D_r$ ) occurs (i.e.,  $DWC > 0.01 \text{ g m}^{-3}$ ) for all flights as a function of estimated  $N_a$ . The  $D_r$  is taken as the cloud depth for ice initiation ( $D_i$ ) if ice particles are evident in the CCP-CIP images. Here, the  $D_i$  is visually ascribed for sizes greater than  $\sim 0.25$  mm and it does not mean that frozen smaller particles cannot be present.”

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## Referee number 2

Report 2:

R2: This is a second round of review process for me. I don't want to review this paper again if further corrections are needed. I feel that this paper needs to be better organized and data analysis should be done properly. I still see that my main concerns are not corrected properly, and text is not improved. Authors insist to keep text same as before after making some minor corrections. I will not go over again my points here but emphasized some points below:

Mainly figures do not represent what is said in the text.

Data analysis does not reflect proper averaging times

A: Averaging times were dictated by the length of cloud passes.

R2: Comparisons with adiabatic calculations were not discussed properly.

A: More explanations is added now at section 3.2 to clarify what is done.

**R2: Title says: Further evidence for CCN aerosol concentrations determining the height of warm rain and ice initiation in convective clouds over the Amazon basin.**

I don't believe that only CCN determines height of warm rain and ice initiation processes.... This is a misleading title, in fact IN or both CCN and IN at high levels plays an important role and never mentioned.

A: The main point of the manuscript is that Na affects Dr. Na is determined mainly by CCN and cloud base updraft. This is shown at Braga et al. (2017).

CCN do affect ice initiation much more strongly than IN in by determining the extent of the Hallett Mossop ice multiplication processes. This is stated in the manuscript in the Introduction, see the text below:

“Ice multiplication is an important mechanism that masks the primary ice nucleation activity when cloud droplets are sufficiently large to promote also warm rain by coalescence, at the temperatures of -3 to -8 °C (Hallet and Mossop, 1974).”

R2: How can adiabatic calculations be made for ref for comparisons or LWC, and used for comparisons if non-adiabatic terms are clearly dominant in a convective process at the certain phases of the storm??? At least 10 times adiabatic values are used for comparisons, in fact, convective clouds may deviate significantly from an adiabatic assumption.

A: The reviewer ignored the large body of literature, described and referenced in the Introduction of the manuscript, showing that the predominant inhomogeneous mixing leads to nearly adiabatic effective radius while LWC can still vary greatly below adiabatic values in the very same cloud volume.

R2: Figures asked to be generated or removed are not included or deleted.

A: We have excluded in the new version Figures 3 and 4 as it was requested.

R2: Fig. 1b is conceptually wrong and not true, if this is the case, what argument you have in the text. I feel this is pointed out previously.

A: But Fig. 1 describes, as well as an illustration can capture, the way that we did the flights and the clouds penetrations. This statement of the reviewer accentuates the notion that he has misconceptions with respect to what the paper is all about.

R2: Fits are provided may not represent data points distribution properly.

A: The fits were generated objectively and show all the data points encountered within the cloud passes during the vertical profiling parts of the flights.

R2: Icing detector and RH plots as indicated before are needed, without them you cant really say particles as droplets or ice crystals when particles are not falling above.

A: Why should RH be relevant within a supercooled water cloud?

The reviewer appears to have ignored the following text in the manuscript:

"The hydrometeor type is identified visually by their shapes. The phase of the smaller CCP-CIP particles cannot be distinguished. Therefore, the precipitation is considered as mixed phase when ice particles are identified, and the combined DWC and RWC are redefined as mixed phase water content (MPWC)."

R2: Images do not show statistically significant data points for droplets or rains. How do you know they are liquid?

A: It is sufficient to identify visually a single ice particle for determining ice initiation. This was done for particles with sizes  $> \sim 0.25$  mm (precipitating particles).

R2: Some text related to CDP and/or CIP probe are not correct.

How collisions and coalescence processes affect cloud macro and micro-properties, how about turbulence?

A: We have no information to address this question. But we can state that most variability in  $D_r$  is explained by  $N_a$ , leaving less room for the impacts of these additional factors. The variability in the relationships between  $N_a$  and  $D_r$  is quantified now and shown at Figure 15.



R2: Conclusions are not provided properly and explained, this was mentioned in my previous review.

I like to see not only  $N_a$  characteristics affecting cloud properties but other parameters such as updrafts and mixing as well radiative processes. Conclusions provided are like for a conference paper.

A: These measurements are not available for the full cloud columns. The fact that  $N_a$  explains much of the variability in  $D_r$  means that there is little room left for all these other processes. The reviewer does not accept this as a possibility. These results found and shown in the manuscript put the relatively small magnitude of these processes in the correct perspective, at least for the Amazon and other regions as Israel and India (shown at Freud and Rosenfeld, 2012). We add the text below in the manuscript.

“The linear relationship between  $N_a$  and  $D_{I3}$  indicates a regression slope of about  $5 \text{ m (cm}^{-3}\text{)}^{-1}$  between  $D_{I3}$  and the calculated  $N_a$  for the Amazon during the dry-to-wet season. This value is slightly larger than the values observed by Freud and Rosenfeld (2012) for other locations around the globe (e.g., India and Israel). These clear linear relationships found between  $N_a$  and  $D_r$  ( $\sim D_{I3}$ ) for different regions highlight the efficiency of the adiabatic parcel model to estimate the height of rain initiation within convective clouds. Additional cloud processes associated such as GCCN, cloud and mixing with ambient air and other processes that are not accounted for in this study produce deviations which are already included within the observed uncertainty of the linear relationship  $N_a$ - $D_r$ ..”

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### Referee number 3

#### Report 3:

R3: This manuscript is a revised version of a previous submission. Although the content has been somewhat improved from the original version, it is still not ready to be released for publication until most of my previous comments are better addressed than they were in the authors' original response. In addition, upon further evaluation of the paper, I have found other sections that need further clarification or modification.

To put my concerns about this paper in context, I will use this sentence in the introduction to highlight what I think is a potentially hazardous approach by modelers: ***“These parameterizations need to represent in simplified form the complex chain of events that occur in clouds.”*** I understand the motivation for parameterizations of cloud microphysical processes in global scale models, and depending on the application of the parameterizations, simplifications can be implemented without seriously misrepresenting the cloud microphysics. In other cases, however, oversimplifications will produce results that are erroneous and misleading. Perhaps it was the first author's wording, and doesn't reflect the thinking of the other authors. If that is the case, then just remove this sentence because I doubt that any serious modeler would agree that the complex chain of events that occur in clouds can ever be represented by simplified parameterization.

A: The sentence is removed. However, much of the point of this paper is that the relationships between cloud base updraft, CCN, Na, Re and Dr obey rather simple rules that are suitable for parameterization. All three reviewers have problems in accepting that other processes such as mixing, turbulence, additional drop activation and even updrafts well above cloud base play a secondary role. This is exactly what makes this paper so important scientifically! If the claim of the paper is really true, it is important news, at least for the reviewers, and more likely to the readers. This does not mean that these other processes are not important. But at least in the clouds that we sampled

their variability from cloud to cloud apparently was not sufficiently large to dominate the relationships between CCN,  $N_a$ ,  $R_e$  and  $D_r$ . We add the text below in the manuscript.

“The linear relationship between  $N_a$  and  $D_{I3}$  indicates a regression slope of about  $5 \text{ m (cm}^{-3}\text{)}^{-1}$  between  $D_{I3}$  and the calculated  $N_a$  for the Amazon during the dry-to-wet season. This value is slightly larger than the values observed by Freud and Rosenfeld (2012) for other locations around the globe (e.g., India and Israel). These clear linear relationships found between  $N_a$  and  $D_r$  ( $\sim D_{I3}$ ) for different regions highlight the efficiency of the adiabatic parcel model to estimate the height of rain initiation within convective clouds. Additional cloud processes associated such as GCCN, cloud and mixing with ambient air and other processes that are not accounted for in this study produce deviations which are already included within the observed uncertainty of the linear relationship  $N_a$ - $D_r$ .”

R3: What further worries me is that the results of this study, as currently presented, suggest that the height of precipitation initiation in any convective cloud can be represented by a single, integer multiplier of the cloud base droplet concentration. In the paper's current form, there are a limited number of caveats to this statement given in the abstract and the summary and yet the caveats are significant and need to be listed just as boldly as the simplified relationship between precipitation initiation height and cloud base droplet concentration.

A: The message that we get from the reviewer is that we should explicitly point out all the competing processes that could affect the  $N_a$ - $D_r$  relationships and discuss how they would potentially affect these relationships. A crucial part of the discussion would be the fact that at the bottom line most variability in  $D_r$  was explained by  $N_d$  (we quantify it better in the new version), thus leaving less room for these other processes. The previous response also addresses this point.

In addition, we have highlighted in the new version of the manuscript the uncertainty regarding the  $N_a$ - $D_r$  relationships. The uncertainty of the linear relationship is mentioned at abstract and summary and is in mean terms of about 21 %, but it does not mask the linear relationship which was found also at different regions around the globe (e.g. Israel and India).

R3: The motivation for forcing such a relationship is not obvious in the current paper but in reading the paper by Freud and Rosenfeld (2012), it seems that the driving force behind these parameterizations is to extract more information from satellite data about cloud properties. This is a worthy objective but not if accomplished at the cost of diminished scientific robustness.

In my comments below, I will further highlight and provide details of where this paper will need to be improved before I will accept it for publication. The bottom line is that I can't and will not allow the publication of a parameterization that can so easily be misused until it is properly justified.

### **Editorial comments**

In my opinion it is my responsibility as the reviewer, a responsibility that I take very seriously, to help improve the paper I am reviewing by 1) identifying technical and factual errors or omissions,

2) requesting clarification when needed and 3) suggesting modifications that help to solidify the hypothesis put forward. In my follow-up review of the revised manuscript, I annotate my remarks with RR1, RR2 or RR3 so that the authors understand the motivation for my comments and suggestions in relationship to how I view my reviewer responsibility (RR).

The authors have sufficiently addressed my concern about the coincidence error losses but all my other comments lack responses that adequately address my comments or concerns. Giving the authors the benefit of the doubt, I will assume the responsibility for not having stated clearly enough the nature of my concerns. Hence I will repeat them here, but with enough detail that there should not be any confusion as to the nature of my comment and how I expect it to be addressed. I will also be more clear about the seriousness of my comments, i.e. those for which I expect concrete changes to the manuscript and those where I will accept lesser modifications as long as my comments receive a reasonable and scientifically defensible response. My comments are not necessarily in the same order as they were presented in the first review.

### **Specific Comments, Questions and Suggestions**

## 1) Error propagation

R3: In Braga et al. (2016) and relatively comprehensive uncertainty analysis is conducted of the number concentrations and sizing by the light scattering and imaging probes; however, this analysis is not taken into account in the current paper to estimate the expected uncertainty in determining Na, Re, Rea, CLWC, Mv, etc. This is a major omission (RR1) that must be rectified.

In their original response to my request to propagate the measurement uncertainties into the derivation of the Dr vs Na relationship, they state:

***“A: The uncertainty of Na calculation with CDP (14 %) is now included in the linear relationship. The linear relationship including Na uncertainty is  $D_r=(5\pm0.7)\cdot Na$ ” .***

A: We have now performed the error propagation where possible. This is shown at Appendix A and Figure 15 in the new manuscript.

This is inadequate since it does not take into account the very large variations in the CLWCa vs Mv relationship that was shown in the Braga et al (2016) paper (Fig. 14a, redrawn below for a single flight), nor does it explain that an additional 30% unexplained correction has been applied to the Na as also explained by Braga et al. (2016), i.e. “However, this methodology does not account for cloud mixing losses from droplet evaporation, and the Na estimates commonly overestimate the expected Nd by 30% (Freud et al., 2011). Therefore, in calculating Na we applied this 30% correction.”.

A: The reason that  $r_e$  can remain near adiabatic while CLWC can vary greatly in the same cloud volume is explained in Section 3.2. We have added the explanation for the 30% correction in Na (Freud et al., 2011) in section 3.2.

R3: The Mv will have at least an uncertainty of 50% since it is derived from rv (I think, although nowhere is Mv ever explained how it is derived). In Fig. 14a, the best fit doesn't even go through most of the points at low or high LWCa so how can a concentration be derived better than to the nearest 100 cm<sup>-3</sup>, much less to the nearest

10th! The supplementary material needs to show the same type of figure as Fig. 14 for all flights so that we can actually see how much deviation there is.

A: Yes,  $M_v$  is the mass of a water sphere having the radius  $R_v$ . This is explained in section 3.2 as follow:

“The uncertainties calculations of cloud properties estimated from cloud probes were described in Braga et al. (2017). The uncertainties of  $r_e$ ,  $r_v$ ,  $r_{ea}$ ,  $r_{va}$  are about 10%, while for CWC and  $M_v$  the uncertainties are about 30%.”

R3: The uncertainty in  $N_a$  then propagates into the  $r_{ea}$  and the derived  $r_e$  will have an uncertainty of more than 20% if the uncertainties in size are properly propagated.

A: We have recalculated the uncertainty of the retrieved  $N_a$ . The uncertainty of  $N_a$  is ~21% in mean terms. The calculation is shown at Appendix A and the values at Table 3.

R3: Finally, there is a very large uncertainty in the  $D_r$ , not only in its derivation of the DWC but also in the actual cloud depth where it is measured. There is an uncertainty of at least a maximum of  $D_r - D_{r-1}$  where  $D_{r-1}$  is the cloud depth at the previous cloud penetration where the  $r_{13}$  threshold was not exceeded. This is because the  $r_e$  threshold could have been exceeded in the rising air mass at any point between the current and previous cloud penetration. This is not addressed at all and is a serious omission.

A: We marked the interval of  $D_r - D_{r-1}$  as the uncertainty for  $D_r$  at Table 3. The uncertainty in  $r_e$  does not change  $D_r$  and  $D_{r-1}$ .

R3: In summary, the  $\pm 0.7$  value is unsupported and requires a much more robust derivation than is currently given.

A: We have recalculated the  $N_a - D_r$  relationship with the error propagation. The results is shown at Figure 15 and Table 3 for each flight. The uncertainty of  $N_a$  is ~21% in mean terms and the linear relationship of  $N_a - D_r$  is  $D_r = (5 \pm 1.06) \cdot N_a$ .

R3: 2. Data processing (RR1)

In my previous review I stated: “In the images from the CIP, there are many out of focus droplets (donuts). The Korolev (2007) correction has to be done, otherwise the derived water content will be an overestimate and the height of precipitation might be incorrect.” To which the authors replied: ***“A: For the data processing of the CCP measurements, ice was assumed as the predominant particle phase in the mixed-state cloud conditions that were mainly given throughout the ACRIDICON CHUVA campaign. The ice assumption causes all images of droplets and ice particles to be treated and considered as particles (apart from shattering-induced particles) but the Poisson spot correction is then excluded. The Korolev correction is defined for liquid drops only and the SODA image processing disables this correction process once the ice-phase is selected. The assumption of ice density instead of water density implies a slight overestimation (~10 %) of the calculated rain water content for particles greater than 75 μm. This will be highlighted in the manuscript.”***

I understand the issue of mixed phase, however, there are three flights, AC09, AC18 and AC19, that have a  $D_r$  that is obviously derived from all water images, so clearly the out-of-focus correction can and should be done. In addition, 2.2.1, it is stated “In this study, we deduced the existence of ice from the occurrence of visually non-spherical shapes of the shadows.” Hence, water droplets can be detected in even mixed phase conditions so that these spherical images can be corrected to derive the true DWC. To not do so will bias the derived  $D_r$  and the subsequent rain initiation level. (RR1).

A: The donuts in the CIP images cannot be removed. But since they are particles that are out of focus this does not matter, as we get the quantitative information from the particles that are in focus. Per the reviewer's request, we recalculated the DWC with CCP-CIP assuming water as particle density for flights AC09, AC18 and AC19. The calculated  $D_r$  did not change, but the DWC is smaller by about ~10-15% when assuming water density instead of ice density. This is presented in section 3.1, as shown in the new text:

“Table 3 shows the cloud depth above cloud base at which warm rain initiation ( $D_r$ ) occurs (i.e.,  $DWC > 0.01 \text{ g m}^{-3}$ ) for all flights as a function of estimated  $N_a$ . The  $D_r$  is taken as the cloud depth for ice initiation ( $D_i$ ) if ice particles are evident in the CCP-CIP images. Here, the  $D_i$  is visually ascribed for sizes greater than  $\sim 0.25 \text{ mm}$  and it does not mean that frozen smaller particles cannot be present. The assumption of water or ice density as the predominant particle phase on DWC calculation based on CCP-CIP probe did not impact  $D_r$  and  $D_i$  measured because the DWC threshold (i.e.,  $DWC > 0.01 \text{ g m}^{-3}$ ) for warm rain or ice initiation was achieved at the same cloud depth for both particles densities. Additional details about the cloud profiling characteristics for each flight as the number of altitude levels sampled (NLS), highest cloud depth without raindrop ( $D_{r,i}$ ) or ice particles ( $D_{i,i}$ ) etc. are also available in Table 3. Furthermore, Appendix A discusses the uncertainty calculations of the estimated parameters of cloud properties.”

### R3: 3. Comparisons of CDP vs CAS-POL and Re vs Rv (RR2 and RR3))

I will reiterate my suggestion that the CDP vs CAS-POL comparisons in Figs. 3 and 4 be removed. First of all, Braga et al. (2016) have more than shown how well these two instruments compare and secondly, there is no reason given in the text of the purpose of comparing Re vs Rv. Freud and Rosenfeld discuss this but it has no relevance in the current paper. These figures and the associated descriptive text should be removed.

A: We have removed Figures 3 and 4.

### R3: 4. Further clarification of the derivation of $N_a$ (RR2)

After reading Freud and Rosenfeld (2012) I was able to decipher the rather cryptic discussion of how  $N_a$  is derived; however, the response to my original request for clarification raises a number of additional questions and related concerns. Here is the original response to my request for clarification:

***“A: The reviewer wrote: “I understand that  $LWC = Nd \cdot Mv$ ”. This is not quite so. The right expression is  $LWC_a = N d_a \cdot M v_a$ , where all are the adiabatic values. The whole idea of the methodology is that the actual  $r_e$  is similar to  $r_{ea}$  - the adiabatic effective radius, due to the nearly inhomogeneous nature of the mixing. The mixing does decorrelate LWC strongly from  $LWC_a$ , while keeping  $r_e$  well correlated with  $r_{ea}$ . The methodology which use LWC vs.  $Mv$  relationship with height to estimate  $N_a$  is well tested and validated at Freud and Rosenfeld (2011). The  $N_a$  estimate is also explained and tested at Braga et al. (2016). Indeed there***



*are uncertainties related to  $N_a$  estimated mostly related when secondary nucleation takes place. The model does not predict that  $N_d$  increases with height, but decrease due to coalescence and inhomogeneous cloud mixing. The results suggest the occurrence of secondary activation with different strengths during flights AC08, AC12, AC13 and AC20 (see figures attached). Large updrafts were measured above cloud base during these flights which increase supersaturation inducing secondary activation.*

This response seems contrary to lines 203: “b) The  $N_a$  at cloud base is estimated through the vertical profile of  $r_e$ .” and line 245: “The  $N_a$  for the convective clusters is estimated based on the slope between the calculated CWC and the mean volume droplet( $M_v$ )”. This seems to indicate that the  $N_a$  is being derived from measurements not the adiabatic values.

A:  $N_a$  at cloud base is indeed estimated through the vertical profile of  $R_e$ , which is expected to be close to  $R_{ea}$ . This vertical profile is measured in a cluster of adjacent clouds. Indeed, additional activation reduces the actual  $R_e$  compared to  $R_{ea}$  and thus induces a positive bias in the computed  $N_a$ . We address it in the new version at section 3.2 as follow:

“The  $N_a$  for the convective clusters is estimated based on the slope between the calculated adiabatic CWC ( $CWC_a$ ) and the mean volume mass of the droplets ( $M_v$ ), which is the mass of a water sphere having the radius  $r_v$ .  $M_v$  is calculated for 1-s DSD measurements of CAS-DPOL and CCP-CDP for non-precipitating cloud passes (Braga et al., 2017). The underlying assumption is that the measured  $r_v$  is approximating the adiabatic  $r_v$  ( $r_{va}$ ) due to the nearly inhomogeneous mixing behavior of the clouds with the ambient air (Beals et al., 2015). Therefore, the measured  $M_v$  approximates the adiabatic  $M_v$  ( $M_{va}$ , where  $M_{va} = CWC_a / N_a$ ).”

R3: This seems to indicate that the  $N_a$  is being derived from measurements not the adiabatic values. In addition,  $M_v$  is never defined, although I was finally able to deduce that it is somehow being derived from  $R_v$ , but this is never made clear.

This response also raises the issue of how adiabatic LWC and  $R_e$  are derived, i.e. one must know not only the cloud base temperature and pressure, but vertical profiles of temperature, pressure and mixing ratio, as well. What vertical profiles are being used? This needs to be described at the very beginning, as well as the uncertainties involved,

i.e. how much do these vertical profiles change over time, especially during the time period of the measurements from cloud base to cloud tops?

A: Since these are convective clouds, we assume simply a moist adiabatic lapse rate within the clouds.

R3: Then the comment regarding “Secondary nucleation” brought forward one of my other critiques regarding of the use of this term. Here is my previous comment:

“9) Secondary nucleation is a very poor term because in a classical parcel model in an updraft, new particle nucleation occurs above cloud base until there are no more cloud active CCN at the level of SS. The implication here is that new CCN are being entrained and that is why the Nd increases with altitude, but this is likely not the case. When running a parcel model with a prescribed updraft and CCN spectra, the supersaturation increases in altitude as the parcel rises adiabatically and cools. The CCN will activate depending on their SS spectra and the available water. This needs revising.”

And this was the response:

***“A: The secondary CCN activation was observed mainly in cloud segments with updrafts that were much stronger than at cloud base. This supports the narrow definition of secondary activation as defined by the reviewer. However, we do not exclude the possibility of additional CCN being entrained and activated above cloud base.”***

This response suggests that the authors did not understand my criticism, i.e. that I did not want them to use the term “Secondary Nucleation” or “Secondary Activation” ANYWHERE in this manuscript for the reasons I stated. They are free to use the term “Additional activation/nucleation” or “Continuing activation/nucleation” but not “Secondary”. If they wish to explain the possibility of entrainment of CCN above the cloud base, they are free to do so, as long as this would be described as “additional” not “secondary”

A: We have corrected the term to additional CCN activation in the whole text.

### R3:5. Precipitation particles coming from below the measurement altitude (RR1)

This concerns a comment I presented above and the response of the authors to a similar comment I had made in my previous review. That comment:

“7) Nothing is said about the uncertainty in the determination of level of precipitation wrt to vertical motions and where the precipitation actually initiated, i.e. it could have actually been below the level of measurement before being lofted upwards. This uncertainty can be estimated using the measured vertical motions.”

The authors' response: ***“A: Doing that would require information that we don't have about the rate of rain formation with height, and will constitute a circular argumentation. The scatter in Figure 17 is the best that we can do for illustrating the uncertainty.”***

This response did not address my concern. In my comments above about the uncertainty in  $D_r$ , at the least, the maximum uncertainty can be estimated as the distance between the two measurement levels with and without the threshold being exceeded, e.g. if the  $R_e$  and  $DWC$  had not been exceeded at the 3000 m level but exceeded at the 4000 m level, given that it might have been exceeded at the 3001 m level that wasn't measured, the uncertainty in this case would be 1000 m. Hence, Table 3 has to have uncertainty bars on ALL of the quantities listed, including the  $N_a$ .

A: We have included uncertainty range on all the quantities in Table 3 and Figure 15.

### R3: 6. Information on time between flight legs through clouds (RR3)

My previous comment:

“8) Nothing is said about the time it takes to make the measurements at the various cloud levels and how these levels were selected. This will give some idea of the time during which the cloud is growing and how long it took to initiate precipitation.”

The authors' response:

***“A: Since the measurements were not following individual growing cloud towers, these times would not advance such knowledge.”***

First of all, the first part of the response seems to fly in the face of what is written on line 126: “The aircraft obtained a composite vertical profile by penetrating young and rising convective elements, typically some 100-300 m below their tops.”

A: We don't see where is the problem. We assume that the time of making the vertical profile (less than an hour) is smaller than the time for changes in cloud base temperature, pressure and CCN.

R3: Secondly, I think that the amount of time from cloud base to each flight level is very germane to the question of how long it takes to initiate precipitation, given the discussion early on concerning the rate at which precipitation forms.

A: The time of cloud growth from cloud base to each level might be important, but the time of aircraft ascent is irrelevant to the cloud processes.

R3: Hence, in Table III I want to see a column that include the time after cloud base measurements that precipitation was identified.

A: We can't see how it would be relevant, because cloud elements that reach precipitating threshold usually already exist at the time that cloud base is measured. Please see Figure 1.

R3: In this same Table I want to see vertical velocity added to the cloud base conditions, another column with the number of levels that were sampled for this date and another column that shows the maximum vertical velocity.

A: We have added  $W_{max}$  and the number of levels that were sampled to Table 3. All vertical velocities from cloud base to the last penetration height are provided at the supplementary material.

R3: This latter request is because it seems to me that from the Supplementary material, almost every flight had vertical velocities above cloud base that were larger than those

at cloud base. This would invalidate the assumptions that are needed to use  $N_a$  as a predictor of  $D_r$ .

A: For additional CCN activation to occur well above cloud base the updraft should be MUCH higher than at cloud base, because of the existence of  $S_{max}$  there. Therefore, having vertical acceleration of the updraft does not mean always additional CCN activation.

### R3: 7. Additional comments

In going through the revised manuscript, I had some remaining questions/comments:

Line 55: The authors use  $-36^\circ\text{C}$  as the threshold for homogeneous freezing but  $-38^\circ\text{C}$  is the value that is most commonly used, hence  $-36^\circ\text{C}$  should be changed to  $-37^\circ\text{C}$ , according to how they state this threshold.

A: We have changed it to  $-37^\circ\text{C}$ .

R3: Line 105: "The  $N_a$  is calculated from  $N_a = CWC_a/M_{va}$ , where  $CWC_a$  is the adiabatic cloud water content ( $CWC_a$ ) as calculated from cloud base pressure and temperature, and  $M_{va}$  is the adiabatic mean volume droplet mass, as approximated from the actually measured mean volume droplet mass ( $M_v$ ) by the cloud probe DSDs obtained during the cloud profiling measurements."

This further confuses me as it implies a mixture of adiabatic and observed quantities, i.e. why is  $M_{va}$  being approximated from the measured mean volume droplet mass, and in addition, why is  $M_v$  never explicitly defined in an equation?

A: The underlying assumption, which will now be stated explicitly, is that the measured  $R_v$  is approximating  $R_{va}$  due to the nearly inhomogeneous mixing. Therefore, the measured  $M_v$  approximates  $M_{va}$ .

The reviewer gathered correctly what is  $M_v$  and  $M_{va}$  (the mass of droplet having radius  $R_v$  and  $R_{va}$ , respectively).

A better explanation is provided at the new version at section 3.2. See below the new text in the new version.

“The  $N_a$  for the convective clusters is estimated based on the slope between the calculated adiabatic CWC ( $CWC_a$ ) and the mean volume mass of the droplets ( $M_v$ ), which is the mass of a water sphere having the radius  $r_v$ .  $M_v$  is calculated for 1-s DSD measurements of CAS-DPOL and CCP-CDP for non-precipitating cloud passes (Braga et al., 2017). The underlying assumption is that the measured  $r_v$  is approximating the adiabatic  $r_v$  ( $r_{va}$ ) due to the nearly inhomogeneous mixing behavior of the clouds with the ambient air (Beals et al., 2015). Therefore, the measured  $M_v$  approximates the adiabatic  $M_v$  ( $M_{va}$ , where  $M_{va} = CWC_a / N_a$ )...”

R3: Line 143: “The DWC is defined as the mass of the drops integrated over the diameter range of 75–250  $\mu\text{m}$  (Freud and Rosenfeld, 2012).” Does this mean that any mass beyond 250  $\mu\text{m}$  is excluded?”

A: Yes.

R3: Line 275: “The precipitation probability is calculated by integrating the measured DSDs exceeding certain DWC thresholds.”. This statement needs a great deal of clarification. First of all, why are these being called “precipitation probabilities” and secondly, are the DSDs being integrated up to the  $Re$  that produces the threshold DWC? Please be more explicit.

A: DWC is integrated between 75 and 250  $\mu\text{m}$ , as the reviewer quoted above.

We have changed the sentence in the new version as shown below:

“The probability of precipitation is the fraction of 1-Hz in-cloud measurements which exceed certain DWC thresholds (i.e. for  $DWC > 0.01 \text{ g m}^{-3}$ ) as a function of  $r_c$  value.”

R3: Line 295: This statement about the relationship between  $D_c$  and  $Re_a$  needs to be qualified with the caveat that it only holds under certain strict conditions, e.g. maximum updraft at cloud base, no continuing activation of CCN above cloud base, etc.

A: we addressed this point by the following added text:

“The  $N_a$  calculation does not take into account the possibility of new nucleation above cloud base (Freud et al., 2011). Braga et al. (2017) have shown that the assumption of adiabatic growth of droplets via condensation from

cloud base to higher levels within cloud can lead to an overestimation by ~20-30% of the number of droplets at cloud base when calculating  $N_a$  in cases with additional droplet nucleation above cloud base.”

The differences between the estimated  $r_{ea}$  and the measured  $r_e$  as a function of  $D_c$  is highlighted at section 4.2.3 as follow:

“This additional CCN activation leads to smaller  $r_e$ . For flights where additional CCN activation was significant, the differences between the estimated  $r_{ea}$  and the  $r_e$  measurements at same height are larger, because the adiabatic estimation does not consider the additional CCN activation of droplets above cloud base and thus overestimates the observed size.”

In addition, the presence of GCCN also can produce differences in the height predicted by the adiabatic model to the rain initiation. In this case we justify in the manuscript (section 4.2.3) our findings for flight AC19 as follow:

“For the flight in cleanest conditions (AC19), the presence of larger aerosol particles (possibly GCCN from sea spray) below cloud base leads to a faster growth of cloud droplets via condensation with height, and consequently  $r_e$  is smaller than 13  $\mu\text{m}$  (see Figure 9a) for warm rain initiation. A similar decrease of  $r_e$  for rain initiation over ocean was observed by Konwar et al. (2012).”

R3: Line 301: The maximum concentration was 2000  $\text{cm}^{-3}$ , but what Nds were used to compare with the Nas?

A: Nd was not directly compared to Na in this study. This was already done in Braga et al., 2017.

R3: Figure 8: Remove it. There is no relevant information here.

A: This image shows the convective nature of the clouds. We prefer to keep it.

## Discussion

Given that the end objective of this study is to support the Freud and Rosenfeld (2012) parameterization so that it can be used by the satellite community to derive microphysical properties, the discussion needs to be much more clear about the

robustness of the  $D_r$  vs  $N_a$  relationship with recommendations as to when it can be used and when it shouldn't be used.

The caveats that limit the use of the  $D_r$  vs  $N_a$  relationship need to be bullets in the Summary so that future use of this parameterization is not used indiscriminately.

A: We have given now a better explanation about the scientific significance and motivation of this study. In the new version of the manuscript (section 1.1) we highlight the scientific motivation of our study as follow:

#### **“1.1. The scientific motivation for this study**

The *in situ* measurements of cloud properties were collected within convective clouds formed over the Amazon from cloud base up to cloud top above the glaciated level. These measurements provided a unique opportunity to evaluate previous theoretical knowledge about aerosol impacts on convective clouds characteristics over the Amazon. In this study the impact of  $N_a$  (adiabatic cloud drop concentrations) in determining the initiation of rain and ice within convective clouds is evaluated. This is performed through the analysis between the calculated  $N_a$ ,  $D_r$  and  $D_i$  for several different environmental conditions over the Amazon (cloud base updrafts, aerosol concentration, surface cover etc.). The relationship of  $N_a$  and  $D_r$  was previously analyzed for regions of Israel and India where a linear relationship was found ( $D_r \approx 4 \cdot N_a$ ) [Freud and Rosenfeld, 2012]. For Amazon region a similar analysis is performed here also taking in account the impact of  $N_a$  in  $D_r$ . This is the first study which analysis the impact of  $N_a$  on  $D_r$  and  $D_i$  at Amazon region using *in situ* measurements of convective cloud properties. The results obtained from comparisons of  $N_a$  estimates and the measured effective number of droplets nucleated at cloud base ( $N_d^*$ ), shown at Braga et al. (2017) for the same flights in the Amazon region, support the methodology of deriving  $N_a$  based on the rate of  $r_e$  growth with cloud depth, and under the assumption that the entrainment and mixing of air into convective clouds is extremely inhomogeneous. This is important because the characteristics of convective clouds based on  $N_a$  values can be extended in space and time by their application to satellite-calculated  $N_a$  (which is obtained with the same parameterization that has been recently developed from the satellite-retrieved vertical evolution of  $r_e$  in convective clouds) [Rosenfeld et al., 2014b].

Regarding the robustness of  $N_a$ - $D_r$  relationship we better highlight the caveats that can limit the use of the equation provided at section 4.2.3 and at the introduction and summary. These limitations were already shown in the previous responses.



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