Manuscript review
Further evidence for CCN aerosol concentrations determining the height of warm rain and ice initiation in convective clouds over the Amazon basin

Braga et al.

General comments

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.

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. 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

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 $D\_r=(5\pm0.7)\cdot Na$".

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.".

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^{-3}, 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.

The uncertainty in Na 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.

Finally, there is a very large uncertainty in the Dr, 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 re 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.

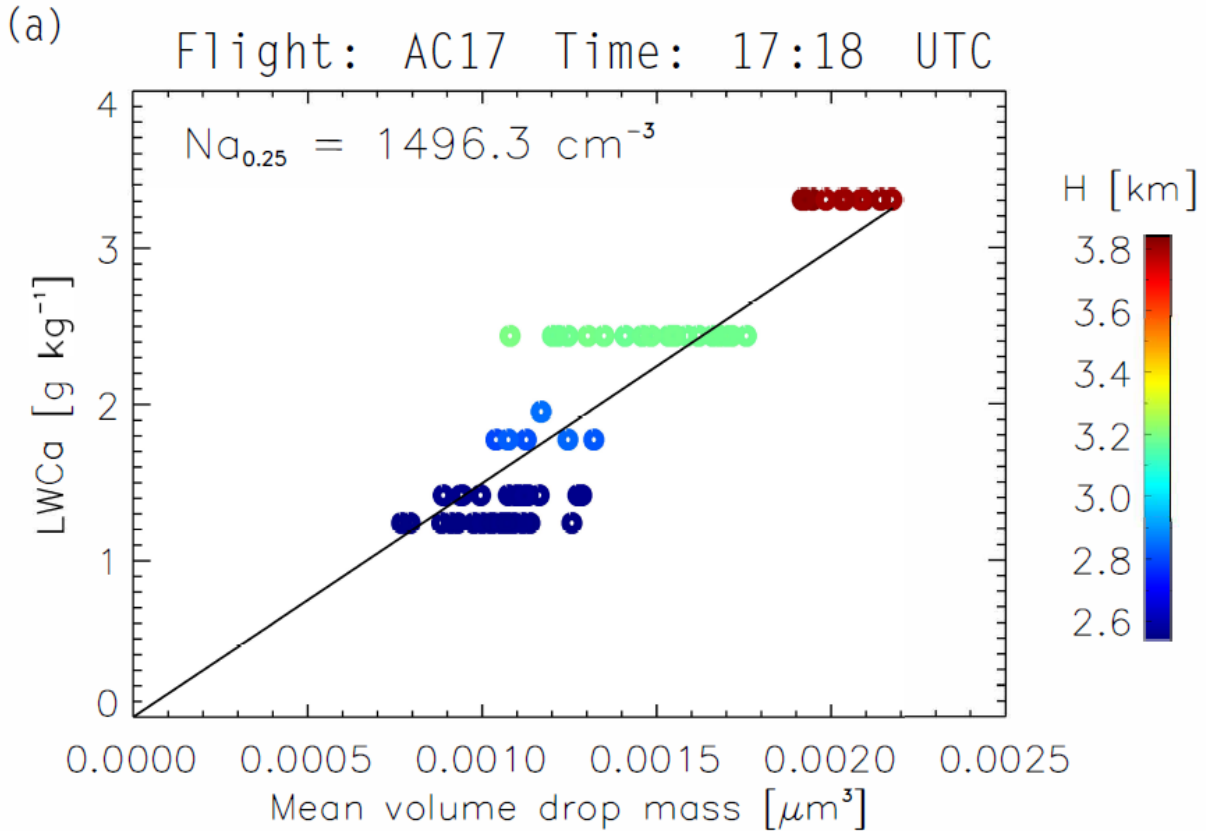In summary, the ±0.7 value is unsupported and requires a much more robust derivation than is currently given.

Figure 14a from Braga et al. (2016)

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 Dr 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 Dr and the subsequent rain initiation level. (RR1).

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.

4. Further clarification of the derivation of Na (RR2)

After reading Freud and Rosenfeld (2012) I was able to decipher the rather cryptic discussion of how Na 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*Mv". This is not quite so. The right expression is LWCa = Nda*Mva, where all are the adiabatic values. The whole idea of the methodology is that the actual re is similar to rea - the adiabatic effective radius, due to the nearly inhomogeneous nature of the mixing. The mixing does decorrelate LWC strongly from LWCa, while keeping re well correlated with rea. The methodology which use LWC vs. Mv relationship with height to estimate Na is well tested and validated at Freud and Rosenfeld (2011). The Na estimate is also explained and tested at Braga et al. (2016). Indeed there are uncertainties related to Na estimated mostly related when secondary nucleation takes place. The model does not predict that Nd 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 *Na* at cloud base is estimated through the vertical profile of *re*." and line 245: "The *Na* for the convective clusters is estimated based on the slope between the calculated CWC and the mean volume droplet(*Mv*)". This seems to indicate that the Na is being derived from measurements not the adiabatic values. In addition, Mv is never

defined, although I was finally able to deduce that it is somehow being derived from Rv, but this is never made clear.

This response also raises the issue of how adiabatic LWC and Re 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?

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 described as "additional" not "secondary"

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 Dr, 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 Re and DWC had not been exceeded at the 3000 m level but is 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 Na.

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 an 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." 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. Hence, in Table III I want to see a column that include the time after cloud base measurements that precipitation was identified. 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. 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 Na as a predictor of Dr.

7. Additional comments

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

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

Line 105: "The *Na* is calculated from *Na= CWCa/Mva*, where*CWCa*is the 105 adiabatic cloud water content (CWCa) as calculated from cloud base pressure and temperature, and *Mva* is the adiabatic mean volume droplet mass, as approximated from the actually measured mean volume droplet mass(*Mv*) 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 Mva being approximated from the measured mean volume droplet mass, and in addition, why is Mv never explicitly defined in an equation?

Line 143: "The DWC is defined as the mass of the drops integrated over the diameter range of 75–250 µm (Freud and Rosenfeld, 2012)." Does this mean that any mass beyond 250 µm is excluded?"

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.

Line 295: This statement about the relationship between Dc and Rea 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.

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

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

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 Dr vs Na relationship with recommendations as to when is can be used and when it shouldn't be used.

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