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Interactive comment on "Aerosol concentrations determine the height of warm rain and ice initiation in convective clouds over the Amazon basin" *by* Ramon Campos Braga et al.

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Interactive response to reviewer's comment on "Aerosol concentrations determine the height of warm rain and ice initiation in convective clouds over the Amazon basin" by Ramon Campos Braga et al. Anonymous Referee #2

The structure is response after the quoted text of the reviewer. The authors thank the referee for the general comments and advices. Furthermore, the advices of the referee 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 reviewer to the paper be considered for publication.

Reviewer's text: A strength of this paper is the presentation of cloud in-situ observations that addresses the question of how convective clouds are influenced by changing the concentration of aerosols. The authors have determined that the height of rain initiation given by D_i is approximately 5 * the number concentration of cloud droplets at cloud base, N_d. This type of study is needed for developing parametrisations. It is difficult to obtain such a wide range of values of N_d using a single location for the study.

There are a number of problems with the paper however.

1. There are no details about the environment of the clouds, the effect of different cloud bases, or the dynamics and history of the clouds. For example, there should be information for each cloud pass about the distance from cloud top, the horizontal distribution along the track of the vertical wind and the location where different measurements were made. Also, it is important to know the history of the cloud before that pass. Does the cloud and the aircraft indeed follow the pattern illustrated in Fig 2? None of the diagrams in Supplementary Fig 1 seem similar to Fig 1b. An example is Figs. 7b-c and 8. How far was the pass below the cloud top? Supercooled raindrops would only be observed in the updraft region.

A: As we mentioned in the manuscript, convective clouds develop as clusters. During the flights, the flight scientist found a region with growing convective cumulus with different stages of development from very shallow to very deep clouds. The cloud passes occurred near the tops of growing convective cumulus were performed about 100-300 m below cloud top, as estimated by the flight scientist, assisted by the pilots (we already highlighted these details while describing Figure 1 at Introduction section). Figure 1 is highly schematic and illustrates the ideas that we measured successively higher clouds near their tops in the same cloud cluster. This type of cloud profile flight was adopted because it provides the closest information as possible about cloud particles formation as a function of cloud depth near the top of growing convective towers. In addition, this type of flight is the safest because the pilots could see the end of the cloud, avoiding the risk of penetrating the cores of Cb. The details about cloud base temperature and heights are available at Table 3. The details about the location of measurements (Figures 1) and vertical velocities (Figures 3) are available at supplementary material.

2. Are there multiple thermals? These can be important for the development of raindrops?

A: No cloud is composed of a single thermal. But the question of multiple thermal is relevant for rain formation only if raindrop that was formed above the cloud penetration level was recirculated to the penetration level. Since we were fairly close to cloud tops, this was not an issue.

3. Turbulence enhancement of collision and coalescence and the enhancement of droplet growth due to entrainment and mixing are not considered in the analysis. Both of these processes would change the simple relationship between D_i and N_d. Giant and ultra-giant aerosols are mentioned, but likewise the analysis does not consider them carefully.

A: The effects of turbulence are inherently fully taken into account in the observed cloud properties, whether we acknowledge it or not. Cloud drop size distribution did not show a separate large mode below the height where cloud drop effective radius > 14 μm, where coalescence leads to fast rain initiation. This means that GCCN did not have a major impact on rain initiation. In any case, as for the turbulence, the reported relationships account for the occurrence of GCCN whether we acknowledge it or not.

4. The initiation of ice particles in convective clouds is complicated. The analysis presented does not discuss the critical aspects of the problem.

A: We just show what we observe. We have insufficient information to go beyond that for ice initiation.

5. The paper is poorly constructed. The font is far too small and diagrams used in discussions are in different documents. Also there are too many similar figures that

show very little and are not properly discussed, while more detailed analysis and accompanying figures are missing.

A: The font used is the required by ACP. We have chosen show many figures at supplementary material to provide the opportunity to the reader check our findings.

Specific comments.

1. Abstract. "Rain initiation" is a loose term. The authors should be more precise."Initiated as ice hydrometeors". The word "initiated" is confusing.

A: Rain initiation is a commonly used term. With respect to ice, the text was changed to: "the first observed precipitation particles were ice hydrometeors".

Does "polluted conditions" include biomass burning?

A: Yes.

Say why smaller cloud droplets froze at lower temperatures compared to the larger [cloud?] droplets in the [un - or less?] polluted cases. And give the sizes.

A: Smaller droplets are less likely to contain immersion freezing ice nuclei due to their lower volume, and are also less likely to meet contact ice nuclei due to their lower surface area. Smaller droplets are also less likely to incur ice multiplication processes. The effective radius of cloud droplets (re) which freezes at -9.1 °C was $\sim$ 11.5 μm for flight AC07, while for flight AC13 re was $\sim$10.2 μm at -14.1 °C. Both flights were performed in polluted conditions over the deforestation arc at Amazon were similar type of aerosols are found (mostly from biomass burning).

"Entrainment and mixing almost completely inhomogeneous". It is the mixing that is either homogeneous or inhomogeneous. A value of r_e close to the r_ea value is not sufficient information to conclude that the mixing process is inhomogeneous. There could be only dilution and no evaporation.

A: The mixing is nearly inhomogeneous.

"Secondary nucleation". It's not true that this process will necessarily inhibit the formation of rain, and may indeed enhance it by providing more small droplets for collisions. Secondary nucleation does not mean that the larger cloud droplets are absent necessarily. Addition of more smaller droplets shifts the value of r_e to smaller sizes.

A: The text says: "Secondary nucleation of droplets on aerosol particles from biomass burning and air pollution reduced re below rea, which further inhibited the formation of raindrops and ice particles and resulted in even higher altitudes for rain and ice initiation." Secondary nucleation slows down the growth with height of the primary nucleated drops at cloud base. Therefore, we support that secondary nucleation prevent the formation of raindrops and ice particles before the first raindrop/ice starts to form. Once new droplets are nucleated above cloud base, the condensational growth rate of the cloud droplets decreases due to the larger competition for the water vapor available. Then, the resulting cloud droplets take more time (requiring thicker cloud depths) to reach larger sizes via condensation process and initiate coagulation or freezing.

2. p2, lines 87-90. It is incorrect to assume that the mixing processes is completely inhomogeneous. And if it was, the vertical profile of the cloud drop effective radius would most likely not follow an idealized adiabatic parcel; there would be broadening.

A: The deviations from complete inhomogeneous mixing explain the deviations shown in many of the figures between the adiabatic and actual re. This is why we claim it is near inhomogeneous, but not ideally so.

3. Line 95. I am not in favour of the wording "raindrops start to form" or "rain initiation" (discussed throughout the paper) since it is a stochastic process.

A: Indeed everything is stochastic, but since the formation rate of raindrops is proportional to the 5th power of re, and re increases with height, it is quite OK to mention height for rain initiation in practical terms. This is what this paper is about.

4. p3, lines 112-113. See #2 above, and furthermore, the vertical values of r_e would

not necessarily be constrained by N_d at cloud base.

A: The previous studies cited here support these characteristics for convective clouds.

5. p3, line 138. Downdrafts can be significant. This is a good reason why the vertical velocity time series and the distance below cloud top should be shown.

A: The reason for limiting the maximum drop size to 250 μm for drizzle water content calculation was because it has a terminal fall velocity of up to 1 m/s. As convective clouds are a turbulent medium large updrafts and downdrafts than 1 m/s were found (this is shown at Figures 3 in supplementary material). Again, this is just an extra measure of caution on top of measuring clouds near their tops, where nothing can come from much greater heights.

6. p6, line 238. What was the wind direction and where were the clouds relative to the opening of the river in Fig1a?

A: The wind direction was east-north-easterly. See the location of the flight in clouds in Figure 1, at the northeastern most edge of the flight track.

7. p8, line 329. Is it possible to show the CDP size distribution just below cloud base? A: Yes. We show in the revised version of the manuscript (Figure 12) the mean total number concentration calculated with PCASP and CCP-CDP for $\sim$200 s of measurements below cloud base during the flights AC12, AC18 and AC19 . The flight AC19 over the Atlantic Ocean (where we indicate the possibility of GCCN) presents higher concentration of particles > 1 μm. This is observed with PCASP and CCP-CDP. The mean total number concentration of large particles measured with CCP-CDP over the ocean is about 10 times greater than observed inland. These values highlight the difference between the size of aerosol which can activate as cloud droplet over the ocean and over Amazon basin.

8. p8, lines 339-340. The adiabatic parcel model would presumably use the aerosol size distribution, including the giant ccn, to initiate the cloud drops.

A: We merely calculated the adiabatic water content and divided by cloud base drop concentrations to obtain adiabatic rv. The re was calculated by re=1.08 rv.

9. p8, line 344. Electrification is not part of this paper. This statement is conjecture.

A: The text was changed from "the low remaining amount of cloud water suppresses the development of cloud electrification" to "the low remaining amount of cloud water reduced a key ingredient for cloud electrification".

10. p8, lines 345-346. It is not evident that downdrafts commence after rain starts. More detailed analysis is needed.

A: Observed downdrafts starts to be evident or more intense above 1660 m after rain starts for flight AC19. Above 1660 m, more downdrafts than updrafts were observed.

11. p8, lines 346-349. The sentence does not make sense.

A: The sentence was rewritten as follows: “The values of vertical velocities measured at flight AC19 (clean region) were smaller than measured for flight AC07 (very polluted region). However, for both cases updrafts are more evident during droplets growth via condensation and downdrafts are most notable when precipitation particles are observed in the cloud.”

12. p8, lines 349-350. More evidence is required. The stronger updraft in Fig 3a of the supplement could be due to environmental conditions.

A: We cannot exclude the enhancement of the updrafts due to environmental conditions. New text: “Strong updrafts ($\sim$10 m s-1) are observed in polluted cases after ice starts to form, probably due to the latent heat release during freezing processes. An alternative explanation of updraft enhancement due to environmental conditions in these cases cannot be excluded.”

13. p9, line 358. There should be more discussion of Figs 13 and 14. Also, again, the images and size distributions should be presented in the context of the updraft

structure. For example, the larger particle in the top panel of Fig 14a looks like a graupel particle. Has particle recognition been performed?

A: The CCP-CIP images were used to distinguish raindrops and ice particles during cloud passes. The hydrometeor type is identified visually by their shapes. The phase of the smaller CCP-CIP particles cannot be distinguished. We believe that at Fig14a we have not graupel particle but some raindrops. The general characteristics of vertical velocities were discussed already in the paper.

14. p9, line 360. As above, it is not right to have some of the figures used in the arguments in the supplementary material and others included in the paper. The diagrams are used in the same way as figures in the manuscript. The authors make a strong statement in the paper based on two cases with one of them shown in the supplementary material.

A: We just highlight what we found. We have already many figures in the paper and some of them we had to send for supplementary material because of editorial objections. We present here this question to the co-editor.

15. p9, line 360 Change the last word in the line (this) to "the" since Fig 5S shows plots constructed for two levels.

A: OK. Thanks.

16. What is the evidence that the images in Fig 5aS are spherical? Some of the larger particles in Fig 5bS do look spherical, but it is not possible to tell for the smaller particles.

A: Indeed, this is why we claim the rain/ice initiation to the larger particles. As we mention before we could not recognize the phase of particles, but only their shapes for precipitable ones.

17. And now back to Fig 14b... Representative images from all parts of the cloud pass should be shown in all cases. What is the difference between almost spherical

particles in Fig 14b and the same in Fig5S? A: Image 14b shows the first pass in which ice hydrometeors are observed mixed with supercooled rain drops. Rain drops were observed also at lower levels, as summarized in Figure 18. Figure 5S shows that in AC18 first rain drops are observed at the -5.7 C isotherm, and that they still remain liquid, or at least spherical, at the -11.4 C isotherm.

18. p9, lines 364 to 371. There is no discussion about the effect of the difference in cloud-base temperature. The main problem with the discussion, however, is there is no mention of the effects of inhomogeneous mixing, or even the decrease in N observed in the two flights: e.g. N_max at 10 deg is the same (500/cc) in AC18 (Fig 4aS) as in AC09 (Fig 13a).

A: The inhomogeneous mixing is implicit when re is closer to rea by the reasons that we mentioned before. At the same temperature Nmax can be the same, but re is larger for AC09, because the 10 C isotherm is higher above cloud base in AC09 compared to AC18.

19. p9, lines 371-374. It is not clear what is meant by the association with vertical velocities? The rate of condensational growth does not depend on vertical velocity. A: The sentence was removed.

20. p9, lines 382 - 384. It is not a relative increase. It is perhaps more surprising that there is such a decrease in N_max between the lowest and next level in AC07 (Fig 5). More analysis should be shown to support suggestions made about secondary activation.

A: We don't assert that it is due to secondary activation. We just raise the possibility.

21. p9, lines 387 - 388. Should it not be the reduced rate of production of raindrops due to lower collision and coalescence efficiencies? There is no process of inhibition or suppression in the cloud.

A: The text was modified to: "These results highlight the role of aerosols in inhibition of

raindrop formation due to inducing a larger Nd and respective lower re, which leads to suppression of collision and coalescence processes in very polluted regions."

22. p9, lines 388 - 389. 300 m is not a great distance when making aircraft passes. Is the result significant? What is the explanation? A: Indeed, it is not a significant difference, but nevertheless in the right direction, this is worth noting.

23. p10, lines 393 - 394. As with so many other statements in the paper, more analysis should be presented to support the statement. What is the variation of CCN and updraft speeds at cloud base, for example?

A: The acceleration of updrafts above the height of cloud base increases supersaturation and thus can induce secondary drop activation. For flights which we observed the increase of Nd with height, high aerosol concentration was observed indicating increased likelihood of secondary nucleation of droplets above cloud base.

24. I believe the discussion and conclusions should be edited based on the referee comments.

A: The conclusions did not change.

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