Authors response to Anonymous Referee #4

Major comments:

1. **(Comment)** This paper used in-situ data from six flights collected during the ACRIDICON-CHUVA field campaign to explore the linkage between gamma distribution parameter phase space and underlying microphysical processes. Three different environmental conditions, the Atlantic Coast, the remote Amazon, and the Arc of Deforestation were studied, and the differences in the underlying microphysical processes among these regions were compared. The paper fits into the scope of ACP and is generally well written, however, the approach used in this study has severe scientific flaws. Therefore, this paper needs to be revised considerably before it can be published in ACP.

1. **(Answer)** We would like to thank Anonymous Referee #4 for the invaluable comments. Please find in this document our detailed answers.

2. **(Comment)** Page 5, Line 15-18. Are there only six flights during the whole field campaign focusing on clouds? If not, why other flights are not used? Especially for Atlantic Coast, there is just one flight used.

2. **(Answer)** Yes, there were other flights that were partially dedicated to probe clouds. However, our specific focus on individual trajectories in this paper meant that we could exchange increased statistics for specialized analysis. The reasoning behind our flight selection was mostly due to the aerosol characteristics below clouds. All flights chosen presented relatively uniform aerosol number concentrations below clouds, therefore avoiding mixture of cleaner and more polluted clouds. On the other cloud profiling flights, we noted variations of the aerosol number concentrations, which would make the analysis much more difficult. By focusing on the selected flights, we were sure that each flight contained almost exclusively the same type of clouds in terms of aerosol conditions. The following sentence was added to the first paragraph in Section 2.1 for clarification: “There were other flights with cloud penetrations, but they are not considered in this study because of higher aerosol variability below clouds. The flights chosen for analysis presented relatively low aerosol variability, meaning that the clouds probed in the same flight were likely subject to similar aerosol conditions”.

There was only one flight over the ocean, so we couldn’t increase its statistics on the paper.

3. **(Comment)** Page 6, Line 22-24: Why PSDs from CIPgs is not used? Only using CDP to create PSDs with D < 50 um will miss out lots of water mass, therefore the third moment used for fitting will be much less.

3. **(Answer)** Our main interest in this study is on the Gamma trajectories of the cloud DSDs. We don’t use CIPgs data because we consider them as drizzle/precipitation DSDs, therefore out of the scope of this study. Besides, there were relatively few DSDs for D > 50 µm. As we explained in our answer #2 to Anonymous Referee #3: “In the warm phase, T > 0 °C, the number of data with $LWC_{D>50} > 0.1$ g m^{-3} is only 8% of the cases where $LWC_{D<50} > 0.1$ g m^{-3} (i.e. only a small portion of the data contained significant amounts of drizzle/precipitation)”.

4. **(Comment)** Page 7, Line 3-14: Incomplete gamma distribution should be used here since only a limited range of particle size is used for fitting. I believe this is the reason why fitted Gamma DSDs are narrower (Page 8, Line 1-4).

4. **(Answer)** While we agree that the incomplete Gamma distribution would fit better the measurements, its use would result in other issues. As one of the main interests of the paper is to study the theoretical Gamma phase-space and its applicability to models in the future, the use of the incomplete Gamma would not be ideal. In a modeling scenario, you don't have the observed DSD and therefore have no way of finding the truncation diameters. Additionally, the use of the incomplete Gamma distribution might add artificial patterns to the phase-space that are due to the truncation and not to physical processes. This point was clarified in the text right before Equation 2 with the following sentence: "The complete Gamma function is used to be consistent with modeling scenarios, where the Gamma parameters are calculated by:".

5. **(Comment)** Section 2.3. I have four major concerns for this method, and will elaborate them in next four points. As stated in Page 9, Line 9, this approach is suitable for the study of the same particle population, which is under Lagrangian framework. Therefore, aircraft dataset at different levels sampling different particle population cannot be used to track the change of cloud PSD gamma parameters, since they are not the same particle population. In addition, the PSDs at the same level are not the same and exhibit large variations. So, the best use of this technique will be for the parcel model if the authors can address the following three comments.

5. **(Answer)** Indeed, this is a valid concern that is also shared by the other reviewers. Here we reproduce our answer #4 to Anonymous Referee #3:

"As you correctly pointed out, it is impossible to produce Lagrangian trajectories based on aircraft microphysical measurements. Therefore, we have to make some assumptions to constrain our method. The flight patterns were specifically chosen in order to follow growing convective elements, where the aircraft penetrated the tops of the clouds. In this way, we both avoided precipitation from above and also tightened the relationship between the altitude of the measurements and the lifecycle of the clouds. We do not presume to claim that this guarantees that our trajectories are Lagrangian. In fact, when it comes to the observations, we never mention it. We just use the altitude of the measurements as a proxy for cloud evolution, meaning that higher measurements present "older" droplets. We believe this is rather reasonable and is also common place in microphysical studies. The confusion might come from the way we described the theory of the Gamma phase space in Section 2.3. In this idealized scenario, we can think of Lagrangian trajectories in order to facilitate the comprehension of the processes that affect DSD evolution. Now the link between the observations and the Lagrangian trajectories, for instance, should be addressed by other means such as modeling. As the title of the paper says, we illustrate the microphysical processes observed in the Gamma phase space rather than attempt to implement it in any actual modeling tool. This is the natural next step, of course, which is already ongoing. We added the following sentence in the second paragraph of Section 2.1 to further detail the flight patterns and why they could be used as a proxy for cloud evolution: "The latter step was deployed as follows. After the cloud base penetration, the aircraft performed several penetrations in vertical steps of several hundred meters. In each step, the aircraft penetrated the cloud tops available, thus avoiding precipitation from above. In this way, different clouds can be penetrated in the same altitude level, but the vertical steps followed the growing cumuli field overall".

In order to compare the results in the paper to a Lagrangian case, we ran a simple model. Please refer to the answer #2 to Anonymous Referee #2. From those calculations, we were able to conclude two things. Firstly, that the qualitative results from the model agree well with our observations. Therefore, even though we could not produce Lagrangian observations, they agree with Lagrangian calculations. Secondly, we were able to test your suggestion regarding the actual calculations of the pseudo-forces (or at least displacements in the phase-space). We were able to confirm the overall directions of the pseudo-forces between the observations and the model, while also quantifying the displacements due to each growth process. The details of the model run can be found in the mentioned answer #2 to Anonymous Referee #2. We also added three new paragraphs to Section 2.3 commenting on the Lagrangian results. Additionally, the answer to Anonymous Referee #2 were also compiled in a new supplement".

6. **(Comment)** Even for the same PSD, there are large uncertainties as stated in Page 6, Line 27-Page 7, Line 1. McFarquhar et al. (2015) studied the uncertainties of counting statistics, and found that all the parameters within an ellipse in Gamma distribution parameter phase space are equally realizable. The displacement of gamma distribution parameters could be just random values in the ellipse unless the ellipse of equally realizable solutions are defined for each PSDs.

6. **(Answer)** Yes, if we consider, for instance, the instrument uncertainty, we would end up with ellipsoids rather than points in the phase-space. However, given that the CDP uncertainty is about 10% (added this information to the second paragraph in Section 2.2), it is clear that the trajectories evolve beyond random movements in an ellipsoid. Let's consider, for the sake of this argument, that the instrument uncertainty is 10% for both the concentrations and the sizing of the droplets. In other words, let's consider that N_d, D_{eff}, and D_g all have 10% uncertainty. Now how that would translate to the phase-space?

We visualize the situation in Figure 10 of the paper. Each uncertainty mentioned could be associated to one of the three axes of the ellipsoids, which are either tangent or normal to the N_d surfaces in Figure 10. Consider that we are in the blue surface of Figure 10 (N_d = 10 cm^{-3}) and in the point where the blue trajectory crosses the upper black line where D_{eff} = 23 μm (they don't actually touch in the Figure 10, but let's consider they do). A 10% uncertainty in N_d would mean a normal axis to the blue surface (both towards and away from the green surface). The size of this axis would be very small in the figure, being 1/100 of the distance between the blue and the green surfaces. The uncertainty in D_{eff} would mean a tangent axis in the direction of the next black line (below), coincidentally of approximately the same size as the distance between the lines. If we consider that a 10% uncertainty in D_g translates to 10% error in ε, then we have the last axis – also tangent to the blue surface, but in the same direction as the D_{eff} line. The projection of the ellipsoid in the blue surface can be roughly represented by the purple curve in the figure below (note that the ellipsoid is very thin in the normal direction). Therefore, the trajectories cover wider regions than the ellipsoids dimensions and the trajectories approach is still valid. We added the following paragraph to the end of Section 2.3 to acknowledge the ellipsoid approach: "Another point to take into consideration are the ellipsoids discussed in McFarquhar et al. (2015). Basically, by considering the instrument and Gamma fitting uncertainties, it is possible to define volumes (with ellipsoid shapes) rather than individual points in the Gamma phase space. Inside each ellipsoid, all DSDs are equally realizable and therefore the movements within it have no particular physical meaning and are statistically the same. In this study, however, we estimate that the results evolve beyond individual ellipsoids and the patterns are associated to physical processes. The results shown in the next sections will not consider the ellipsoid approach, but the points shown can be considered to be the central points of such volumes".

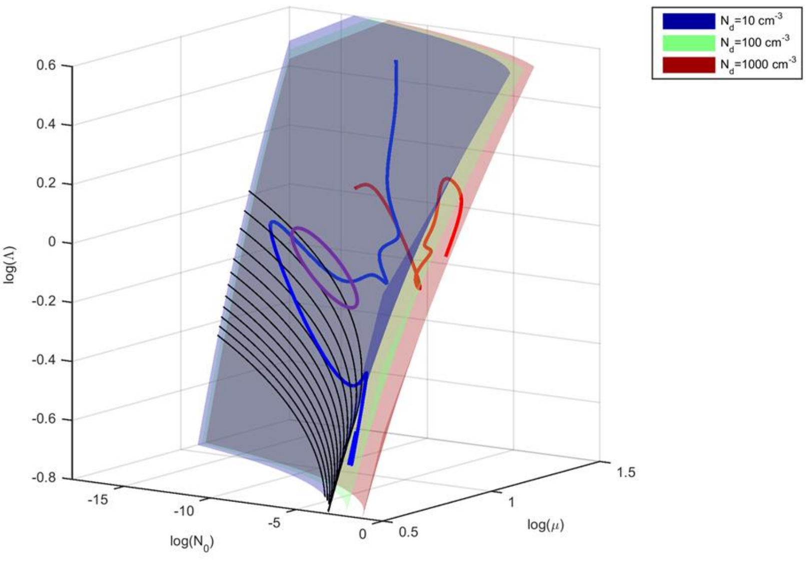

Figure R1: estimated size of the uncertainty ellipsoid in Figure 10 of the paper.

From this simple calculation, we conclude that the trajectories are likely to evolve beyond random movements in one ellipsoid. We won't carry this calculation over to the paper, however, because the trajectories can only be defined by points instead of volumes. In that sense, we can consider that the points in the trajectories are the central points of the ellipsoids. In the future, it would be interesting to study how the ellipsoids can be understood taking into account the underlying physics found in the phase-space.

7. **(Comment)** As for the "pseudo-forces", or microphysical processes which I prefer, this study decomposed it into two components: condensational growth and collision-coalescence growth. Due to the complex microphysical processes occurring in the clouds (as is discussed by the authors in Page 9, Line 21 – Page 10, Line 2), the evolutions of PSDs are very complex as some simulations using bin microphysics show. Simply relating a change of gamma distribution parameters to either condensational growth or collision-coalescence is not justified. Especially for any volume of air the aircraft sampled (or numerical models in Eulerian framework), the horizontal and vertical advection are very important.

7. **(Answer)** Yes, there are several other processes that affect the DSDs. However, we would argue that condensation and collision-coalescence are definitely the most determinant. Any model, however simple, should be able to reproduce those mechanisms in order to explain precipitation formation even though the other processes are also important. These other forces are also being studied in the phase space using different microphysical parameterizations and we hope to have further results soon. For this first introduction of the phase-space, we chose to focus on condensation and collision-coalescence in order to define the overall characteristics of the space and how we can analyze cloud DSDs evolution in it. The other processes can and should be analyzed on top of that basis.

Let's take the mixing processes as an example. There are several studies analyzing the effects of homogeneous or inhomogeneous mixing on the characteristics of the DSDs. Therefore, in principle, we should be able to estimate the effects of mixing on Gamma DSDs as well, which can be reproduced in the phase-space. This could explain some characteristics of the trajectories we observed, but we believe this kind of analysis is beyond the scope of our paper. We are considering this kind of analysis as the necessary next steps, which should involve other tools such as models.

That said, we ran a simple Lagrangian model with bin microphysics in order to check the patterns associated to condensation and collision-coalescence and if they match our observations. If they qualitatively agree, it means that our observations are capturing those microphysical processes. Please refer to our answer #2 to Anonymous Referee #2 where we detail the model runs and its results. Overall, we were able to confirm that the condensation and collision-coalescence processes induce displacements in the same directions that we inferred from the observations. Obviously, the quantitative results are different given that the model is relatively simple and does not consider several processes that affect our observations. But we believe this is a good indication that the Gamma phase-space methodology is consistent and that we should dedicate efforts to progressively include the other processes as well. In that regard, it would be interesting to analyze the Gamma phase-space in more complex models such as the LES-type.

8. **(Comment)** The directions and magnitudes of "condensational growth pseudo-force" and "collision-coalescence pseudo-force" are uncertain, which means that the influences of each individual microphysical processes on PSD evolutions are not studied clearly.

8. **(Answer)** Yes, we didn't explicitly quantify the pseudo-forces in this first introduction of the methodology. The main intent of this paper is to introduce the Gamma phase-space as a physical entity and illustrate microphysical processes in it. We believe the quantifications should be the focus of future implementations of the phase-space and is beyond the scope of the present work. However, the Lagrangian model runs we mentioned in the previous item can be considered as the first step in that direction. Note that the direction of the displacements is similar between the model and our observations. Therefore, the displacements in the observed trajectories can be at least partially explained by the processes considered in the model.

9. **(Comment)** The descriptions of "favors high value of mu while slightly increasing lambda" (Page 10, Line 4) and "lower values of lambda and mu, the former decreasing at a faster pace" (Page 10, Line 13- 14) are not precise and not justified. The change of N_0 as described are wrong, since if condensational growth increase both μ and λ while keeping the same total number concentration, N_0 should also increase. In addition, if collision coalescence lower both μ and λ, and total number concentration of course, then N_0 should be also decreasing. Besides, I would say that evaporation "pseudo-force" acts the opposite way as "condensational growth pseudo-force" instead of "collision-coalescence pseudo-force" in this study. Anyway, the directions of these "pseudo-forces" are totally unknown, and the change of gamma distribution parameters could be any microphysical processes since relating the change of gamma distribution parameters (or equivalently PSD moments or bulk properties) to any single microphysical process is impossible.

9. **(Answer)** While our affirmations may not be precise, because it is pattern analysis instead of actual quantification, they are correct. Both our observations and the model calculations corroborate those affirmations. Please refer to our answer #2 to Anonymous Referee #2. In that document, we calculated the elevation angle ϕ to be positive for the condensational growth, meaning increasing Λ. On the other hand, we calculated an average θ of 179.6 °. Because this angle is calculated from the $\log(N_0)$ axis, this value means growing μ and shrinking N_0 – second quadrant. For collision-coalescence, $\phi = -4.23$ °, meaning decreasing Λ. The azimuth θ is -13.7 °, which is in the direction of growing N_0 and decreasing μ. We believe those quantifications are the first step at calculating the actual values of the pseudo-forces, but its implementation should be the focus of further studies in the future.

The “evaporation pseudo-force” is surely the opposite of the “condensational pseudo-force”. But it is also true that the collision-coalescence pseudo-force acts in the opposite (overall) direction as the condensational pseudo-force.

Minor comments

1. **(Comment)** Page 4, Line 24-25. This sentence needs to be elaborated.

1. **(Answer)** Changed to: “This process may produce artificial trajectories in the phase space by limiting the parameter variability”.

2. **(Comment)** Figure 1. Add flight height and temperatures for each flight.

2. **(Answer)** We believe it would be hard to visualize the altitude and temperature in this figure. Instead, the requested properties can be seen in the other figures and tables in the paper.

3. **(Comment)** Page 5, Line 23 – Page 6, Line 10. The three regions used in section 3.2 should be introduced here clearly. Furthermore, the cloud characteristic for coastal region and remote Amazon should be described here, similar to what has been written for the Arc of Deforestation.

3. **(Answer)** Added the following sentence to the end of the second paragraph in this section: “Contrasting with the Arc of Deforestation, the region named Remote Amazon in this study has much lower background aerosol concentrations, producing cleaner clouds. Clouds over the Atlantic Ocean developed under cleaner conditions as compared to the continental counterparts, and also had lower cloud bases (Table 1)”.

4. **(Comment)** Figure 5-8. The y and z axes (μ and λ) don’t need to be taken logarithm for easy comparisons with previous studies. In addition, the projection of the 3D trajectories in N_0-μ, N_0-λ, μ-λ planes will make readers to follow easier.

4. **(Answer)** We analyzed the situation and decided that logarithmic axes were the easiest way to visualize the trajectories. The projections were added to the figures.

5. **(Comment)** Figure 5-8. Add raw PSDs with different colors showing different time, so the change of PSDs is clear to the readers. As shown in many previous studies (e.g., Heymsfield et al. 2013), the gamma distribution parameters can compensate with each other, therefore, the different points in the gamma distribution parameter phase space could mean the same PSD.

5. **(Answer)** We show observed and fitted DSDs in Figure 9. As far as the trajectories go, we did not observe any pair of similar DSDs with different Gamma parameters.

6. **(Comment)** Page 14, Line 23-27. Recommend removing these sentences. As stated in Major comment #7, the quantitative descriptions of these “pseudo-forces” are lacking. Besides, the method may just work for Lagrangian framework. I cannot see how this could be used for bulk microphysical schemes.

6. **(Answer)** We have shown that it is possible to produce quantifications of the processes using a simple Lagrangian model. Of course, the method should be refined to consider the many other aspects present in clouds. We believe it is important to leave those affirmations in the paper, as they can be addressed by other researchers.

7. **(Comment)** Page 17, Line 10-21. According to Equation 9, this is similar to fix µ which is adopted in lots of numerical schemes. Actually, the small range of µ is due to its scale, and could mean large variations of PSDs.

7. **(Answer)** We left the second sentence in the last paragraph of Section 3.2 more open-ended: "If *ε* can be constrained in the model, it should be possible to obtain the full Gamma DSD – which is the point in the intersection curve that presents the given *ε*".

8. **(Comment)** Page 19, Line 11-12. The sentence that "The characteristics of the clouds warm layer. . .should have a determining role in the glaciation initiation". I would argue that the IN and the ice microphysics above are more important. The characteristic of IN between the remote Amazon and the Arc of Deforestation are not studied. The number concentration of ice particles above should also be analyzed, which may explain the differences in glaciation.

8. **(Answer)** In this sentence we refer to the glaciation initiation – imagining a cloud that is growing past the 0 °C and does not have an ice phase yet. In this scenario, the characteristics of the droplets that cross the 0 °C isotherm are definitely important to trigger (or not) the glaciation process.

Yes, it is unsure how the IN population changes with pollution, but previous studies suggest that most of the IN over the Amazon come from natural sources – either from the forest or from long range transport (Saharan dust).